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An Assessment Model for Atmospheric Composition

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An Assessment Model for Atmospheric Composition

Edited by
Michael J. Prather
NASA Goddard Institute for Space Studies
New York, New York

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PREFACE

This workshop grew from discussions in early 1987 between Dennis Tirpak of the EPA and Michael Prather of NASA/GISS regarding models to predict atmospheric composition. Such models provide one component of EPA's overall program to assess the impact of projected societal actions on global change. Michael Prather agreed to convene a workshop comprising members of the atmospheric sciences community who have been working on theoretical modelling of one or more aspects of atmospheric composition. The objectives were to build a parameterized model and to assess its accuracy in representing the best current research models.

The parameterized model and reviews are documented in this report. Attempts were made to recognize the uncertainty of such predictions by calculating a range of solutions based on upper and lower limits for selected parameters. References are limited, used mainly as pointers to research from which the parameterizations are taken.

Based on the experience of developing the parameterized model and reviewing the final report, the participants have contributed formal, critical reviews that are presented verbatim in the conclusions. It became clear that such models can only be as good as the current research in each of the individual areas and that the models fail to represent adequately the uncertainties in modelling. There were strong feelings that these types of models must be continually updated and maintained by the scientific community to reflect their most recent levels of research. Therefore, the model described here may be regarded as representing an abstracted part of current atmospheric research and cannot be appropriately used in succeeding years.

Michael Prather
NASA/GISS, New York
31 August 1988

PARTICIPANTS, CONTRIBUTORS and REVIEWERS

Dan Albritton	NOAA, Colorado
Robert Dickinson	NCAR, Colorado
Inez Fung	NASA, New York
Richard Gammon	NOAA, Washington
James Holton	U. Wash., Washington
Ivar Isaksen	U. of Oslo, Norway
Malcolm Ko	AER, Massachusetts
Andrew Lacis	NASA, New York
Dan Lashof	EPA, D.C.
Shaw Liu	NOAA, Colorado
Jennifer Logan	Harvard, Massachusetts
Jerry Mahlman	NOAA, New Jersey
Pauline Midgley	ICI, Delaware
Michael Prather	NASA, New York
Ron Prinn	MIT, Massachusetts
Nien Dak Sze	AER, Massachusetts
Anne Thompson	NASA, Maryland
Dennis Tirpak	EPA, D.C.
Don Wuebbles	LLNL, California

1. INTRODUCTION

Predicting future perturbations to global air quality and climate requires, as a prerequisite, prognostic models for the composition of the Earth's atmosphere. Such assessment models are needed to evaluate the impact on our environment of different social choices that affect emissions of photochemically and radiatively important trace gases (see reviews GTC, 1986; WMO, 1986). Our presentation here of a prototype assessment model is intended to encourage public scientific discussion of the necessary components of the model and their interactions, with the recognition that models similar to this will likely be used by the Environmental Protection Agency and other regulatory agencies in order to assess the effect of changes in atmospheric composition on climate over the next century.

As part of this workshop we built and examined a highly parameterized model for calculating atmospheric composition. We found that such a model has advantages in coupling components of chemistry, composition and climate within one model, but it is limited in representing current theoretical models by obscuring the physical processes occurring in the atmosphere and thereby hiding some of the uncertainties.

The assessment model described here predicts the atmospheric concentrations of chemically and radiatively active trace gases through the 21st century, based on an assumed scenario for emission of trace gases. The model is intentionally simplified, but brings together the major chemical species affecting the composition of the stratosphere and troposphere, and hence the radiative forcing of the climate. These species include several long-lived trace gases, ozone, other short-lived chemicals and water vapor. The changes expected in climate and stratospheric ozone are driven primarily by the increasing concentrations of the long-lived trace gases such as CO_2, N_2O, CH_4, H_2O and the CFCs, with some additional, but more uncertain contributions from the radiative coupling with O_3. The model predicts ozone to the extent that it would affect the lifetimes of the other trace gases and includes predictions of temperature to the extent that temperature impacts concentrations of trace gases through chemistry and equilibrium vapor pressures. The predictions for ozone and temperature should not be used independently to replace more detailed and more accurate model studies of these perturbations.

The model is straightforward to program and is computationally inexpensive. Feedbacks between any of these variables in the model can be explicitly included. The advantage of the simplified model is that it combines all the major variables into a single, fully coupled framework so that the sensitivity of climate response to particular forcing can be easily compared. Upper and lower limits placed on all variables are an important part of the model: they are used to stop the calculation when any atmospheric variable exceeds the range for which the approximations used in constructing the parameterizations are no longer valid.

A major disadvantage of this type of proposed model, beyond the obviously limited accuracy, lies in the mathematical, non-physical nature of the parameterizations. This assessment model cannot begin to represent the detailed, spatially and seasonally resolved simulations of the atmosphere currently available from the most advanced research tools available in each discipline: such as, transient-climate calculations with current 3-D general circulation models, or time-dependent scenarios for stratospheric ozone with current 2-D models. Nevertheless, the

dominant effects on the mean global atmosphere from these calculations can often be reduced to first and second-order relationships between the major climate variables. Furthermore, the realistic coupling of these discipline-specific models has not yet been achieved.

A primary assumption of this model for atmospheric composition is that *annually and globally integrated quantities* suffice to define the first-order effects on climate, stratospheric ozone, and tropospheric oxidants *in so far as they control atmospheric composition.* In a few cases we find it necessary to define separate variables for both stratospheric and tropospheric concentrations, or for both hemispheres. These chemical species and related quantities, described in Table 1, are the fundamental variables of the model which are calculated or specified as a function of time in the model. A secondary assumption is that these constituents may be treated as one of three types: (1) those with long lifetimes which can be explicitly integrated over time from a set of initial conditions, (2) those in steady state with their environment because their time scale for adjustment is 1-2 years or less, and (3) those specified *a priori* on the basis of a given scenario.

Integration of any such algebraic model is deterministic, in that there exists one unique solution for a given scenario; however, this single history of atmospheric composition does not reflect uncertainties in the predictions. We present a new approach for representing the uncertainty inherent in model predictions. For a fixed scenario of emissions the model calculates a manifold of solutions, 2^n, representing the possible histories associated with a specified range in each of n key parameters of the model. We chose a value for n of 6 in order to display a sufficient range of results. These efforts constitute neither a formal nor an extensive analysis of uncertainties, and more work is needed in this area.

The components of the model are defined in Table 1. Section 2 describes the treatment of the long-lived trace gases. Climate variables, stratospheric constituents, and the elements of tropospheric chemistry are described in Sections 3, 4 and 5, respectively. The model program is listed in Table 2, the coefficients for the model are given in Table 3, and the input scenario for the test case described in Section 6 is given in Table 4. Figures of the model results, including the manifold of solutions representing a range of uncertainty in the predictions are discussed in Section 6. Section 7 presents critical reviews from the participants and outside reviewers.

Table 1A. EXPLICITLY INTEGRATED SPECIES

			reference state*	
A.	N_2O	X_A	(300 ppb)	(nitrous oxide)
B.	CH_4	X_B	(1600 ppb)	(methane)
C.	CO_2	X_C	(345 ppm)	(carbon dioxide)
D.	$CFCl_3$	X_D	(220 ppt)	(CFC-11)
E.	CF_2Cl_2	X_E	(375 ppt)	(CFC-12)
F.	CHF_2Cl	X_F	(80 ppt)	(CFC-22)
G.	$C_2F_3Cl_3$	X_G	(30 ppt)	(CFC-113)
H.	CCl_4	X_H	(100 ppt)	(carbon tetrachloride)
I.	CH_3CCl_3	X_I	(110 ppt)	(methyl chloroform)
J.	CH_3Cl	X_J	(600 ppt)	(methyl chloride)
K.	CF_3Br	X_K	(1.7 ppt)	(Halon-1301)
L.	CF_2ClBr	X_L	(1.5 ppt)	(Halon-1211)
M.	CH_3Br	X_M	(10 ppt)	(methyl bromide)
N.	CF_4	X_N	(60 ppt)	(carbon tetrafluoride)
T.	trop-T	X_T	(0.5 °C)	(tropospheric temperature: perturbation)

Annual average values of long-lived, slowly changing species, integrated with a one-year time step. All mixing ratios in this paper are vol/vol.

* The reference state refers to "present-day conditions", circa 1985; this state is NOT necessarily the initial state for the calculation. Not enough is known about the pre-industrial atmosphere to allow it to be used as a reference state.

Table 1B. IMPLICITLY SOLVED SPECIES

a.	trop-OH	x_a	(0.0 %)	(global mean = NH + SH)
b.	NH-O_3	x_b	(0.0 %)	(N.H. "mean" tropospheric O_3)
c.	NH-OH	x_c	(0.0 %)	(N.H. "mean" OH: pert. to reference state)
d.	NH-CO	x_d	(100 ppb)	(N.H. annual mean CO concentration)
e.	SH-O_3	x_e	(0.0 %)	(S.H. "mean" tropospheric O_3)
f.	SH-OH	x_f	(0.0 %)	(S.H. "mean" OH: pert. to reference state)
g.	SH-CO	x_g	(60 ppb)	(S.H. annual mean CO concentration)
h.	trop-H_2O	x_h	(0.0 %)	(pert. to "mean" tropospheric water vapor)
i.	col-O_3	x_i	(0.0 %)	(total ozone column)
j.	upp-O_3	x_j	(0.0 %)	(ozone column above 30 km)
k.	str-NO_y	x_k	(18 ppb)	(HNO_3+NO+NO_2+NO_3+2xN_2O_5+HNO_4+$ClNO_3$, ~35km)
l.	str-Cl_x	x_l	(2.78 ppb)	(HCl+Cl+ClO+2xCl_2+HOCl+$ClNO_3$ @ ~40 km)
m.	str-Br_x	x_m	(12.9 ppt)	(BrO+Br+HBr+HOBr+$BrNO_3$ @ ~25 km)
n.	str-H_2O	x_n	(3 ppm)	(@ tropopause)
o.	net-F	x_o	(0.0 W/m^2)	(net radiative forcing: T, gases, albedo)

The annual steady-state values of short-lived variables, whose values are given as absolute concentrations or perturbations (% or W/m^2) to the reference state. net-F is calculated from the radiative forcing relative to an estimated pre-industrial atmosphere (see text).

Table 1C. EXTERNALLY SPECIFIED SCENARIOS

A-N.	fluxes	S_{A-N}	(X g/yr)	(annual emission of long-lived gases)
1.	albedo	S_1	(0%)	(pert. to planetary albedo)
2.	ocean	S_2	(0%)	(pert. to ocean heat/CO_2 capacity)
3.	circ	S_3	(0%)	(pert. to strat. circulation)
4.	NH/CO	S_4	(400 Tg/yr)	(N.H. annual mean CO flux)
5.	SH/CO	S_5	(80 Tg/yr)	(S.H.)
6.	NH/NO_x	S_6	(36 Tg(N)/yr)	(N.H. flux)
7.	SH/NO_x	S_7	(14 Tg(N)/yr)	(S.H. flux)
8.	NH/NMHC	S_8	(1000 Tg/yr)	(N.H. flux)
9.	SH/NMHC	S_9	(500 Tg/yr)	(S.H. flux)

Stratospheric circulation is measured here by the rate of influx of air across the tropical tropopause into the lower stratosphere; it can be measured by the mean residual vertical motions or the net diabatic heating in the lower tropical stratosphere.
Fluxes (CO, NO_x and NMHC) are denoted by a slash (/) and the values quoted here refer to reference state conditions.

Table 1D. FLOW DIAGRAM

(0) Initialize the variables in Table 1A (N_2O, CH_4, ... trop-T) at beginning of year 1985 (time = 0).

(1) Begin with the values for variables in Table 1A (N_2O, CH_4, ... trop-T) at beginning of year 19xx (time = T). These can be taken from the initialization (0, above) or from the last calculation cycle (4, below).

(2) Look up the values for the variables in Table 1C (fluxes for species in Table 1A, albedo, circulations, fluxes for tropospheric CO, NO_x & NMHCs) for year 19xx (time = T to T+1).

(3) Calculate the steady-state values for the dependent variables in Table 1B (col-O_3, ... trop-H_2O) as a function of the variables specified in (1) and (2) above. These values are assumed to be applicable for the entire year 19xx (time = T to T+1).

(4) Use the lifetimes and fluxes calculated in (2) and (3) above to integrate the variables in Table 1A (N_2O, CH_4, ... trop-T) forward to the beginning of a new year 19xx+1 (time = T+1). Go to (1, above).

Table 1E. A SAMPLE EMISSIONS SCENARIO

EPA Scenario (low CH_4, low CO, high NO_x, protocol for CFCs)

species	1985 flux (Gg/yr)	trend (%/yr)	% growth per year in the decade 1980 1989	1990 1999	2000 2009	2010 2019	2020 2029	2030 2039	2040 2049	2050 2059	2060 2069
N2O (N)		(0.25)	1.9	-.1	0.9	0.3	.67	.67	.67	0.8	0.8
CH4		(1.0)	.76	.79	.78	.75	.60	.60	.60	.27	.27
CO2 (C)		(0.4)	1.23	.86	1.28	1.46	1.19	1.19	1.19	1.22	1.22
CFCl3	270.		5.7	-1.5	-0.7	-1.3	0.2	0.5	0.5	0.1	0.1
CF2Cl2	400.		3.8	-0.5	-0.3	-0.4	-1.9	0.3	0.3	0.1	0.0
CHF2Cl	225.		10.1	4.9	3.0	2.5	2.5	2.5	2.5	1.2	0.3
C2F3Cl3	140.		0.5	-4.3	-1.1	0.4	0.4	0.4	0.5	0.5	0.5
CCl4		(2.0)	3.6	-3.5	-1.0	0.3	0.3	0.4	0.4	0.4	0.4
CH3CCl3	520.		-3.3	2.7	2.5	2.5	2.5	2.5	2.5	2.5	2.5
CH3Cl		(0.0)	0.0	...							
CF3Br	6.		18.0	10.3	6.2	1.8	0.6	0.3	0.3	0.3	0.2
CF2ClBr	5.		11.2	2.6	2.3	1.0	0.3	0.2	0.5	0.3	0.1
CH3Br		(0.0)	0.0	...							
CF4		(5.0)	0.0	...							

species	flux (Tg/yr)	trend (%/yr)	1980 1989	1990 1999	2000 2009	2010 2019	2020 2029	2030 2039	2040 2049	2050 2059	2060 2069
albedo		0.	0.0	...							
ocean		0.	0.0	...							
circ		0.	0.0	...							
NH-CO	400.		-2.4	-2.1	-0.7	-0.9	0.0	0.0	0.0	0.0	0.0
SH-CO	90.		0.0	...							
NH-NOx	20.		0.0	0.0	0.0	0.42	0.50	0.50	0.50	0.72	0.72
SH-NOx	10.		0.0	...							
NH-NMHC	30.		0.0	...							
SH-NMHC	10.		0.0	...							

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2. LONG-LIVED TRACE GASES

The long-lived trace gases described in Table 1a are the focus of this model. Atmospheric destruction rates for these gases are conveniently discussed in terms of chemical lifetimes. The *local* chemical lifetime is defined as the concentration divided by the *local* chemical loss rate. For photodissociated species, it depends on ultraviolet fluxes and photolysis cross-sections, and for many oxidized species, on local OH concentrations and temperature. The *global* lifetime for a species against a particular loss is defined as the global content divided by the *globally* summed losses. Uncertainties in local lifetimes exist because the ultraviolet fluxes depend sensitively on the overhead ozone amount and on detailed knowledge of radiative transfer (particularly in the Schumann-Runge band region), and because even a partially validated model for OH concentrations is not yet available. Uncertainties in global lifetimes exist because of the further difficulty in integrating the product of loss rate times species' concentrations over the highly variable global environment.

For the perhalogenated hydrocarbons ($CFCl_3$, CF_2Cl_2, $C_2F_3Cl_3$, CCl_4, CF_3Br, CF_2ClBr) and nitrous oxide (N_2O) we use lifetimes computed assuming stratospheric loss only (Golombek and Prinn, 1986; Connell, 1986; Ko et al., 1985; Wuebbles, 1983). These values are consistent with the fact that the ALE/GAGE measured lifetime for $CFCl_3$ (Cunnold et al., 1986) agrees very well with the theoretical lifetime computed from models assuming stratospheric destruction exclusively. For CH_4, CH_3CCl_3, $CHFCl_2$, CH_3Cl, and CH_3Br the dominant loss is in the troposphere by reaction with OH. Standard, steady-state lifetimes are computed by scaling the ALE/GAGE lifetime for CH_3CCl_3 (Prinn et al., 1987) with the relevant, temperature-dependent rate constants for the OH reactions. This approach gives lifetimes consistent with models for global OH. For the longer lived species, CHF_2Cl and CH_4, stratospheric losses are also considered.

When computing the global lifetimes as needed in this assessment model, the relative spatial distribution of sources and sinks needs to be considered carefully. In this model we specifically utilize the global chemical lifetime defined under steady-state conditions: that is no temporal trends in concentration for a constant source strength. For those long-lived species with predominantly tropospheric losses (CH_4, CH_3CCl_3, etc.) the steady-state lifetimes reflect their instantaneous loss; and thus the annual loss is given by the current global content divided by the current steady-state lifetime in years. For species with predominantly stratospheric sinks ($CFCl_3$, CF_2Cl_2, N_2O, etc.), there will be a significant lag between the buildup of global burdens in the troposphere and the annual stratospheric loss. In this case, the global destruction rate at time t (years) is equated to the global content at time ($t - t_{lag}$) divided by the current steady-state lifetime in years. Here t_{lag} is a time lag associated with transport from the surface source region to the stratospheric destruction region for these species. We use t_{lag} = 2.5 years, that is, the Jan 1, 1990 burden is used to calculate average loss throughout the year 1992.

The global content of a long-lived trace gas (X's in Table 1a) is integrated on an annual basis (Δt = 1 year) by adding the annual source (S's in Table 1c) and removing the annual losses according to the current, global-average, steady-state lifetimes (in years) associated with stratospheric (L_s) and tropospheric sinks (L_t).

$$X(t+1) = X(t) + S(t) - X(t)/L_s(t) - X(t-t_{lag})/L_s(t)$$

Lifetimes in the model are updated annually in response to predicted perturbations in ozone column amounts, stratospheric transport rates, atmospheric temperatures, and OH concentrations. These sensitivity coefficients use linearized relationships based on results from the discipline-oriented research models (see references for the models used by workhsop contributors) and are given in Table 3 (Model Coefficients). Sources of the trace gases are updated annually from the externally imposed scenario (Table 4) which specifies the percent change with respect to the previous year. For gases with significant natural sources or with uncertain anthropogenic components, the initial fluxes are adjusted so that the currently observed rate of increase is consistent with the lifetime.

CO2

Carbon dioxide, CO_2, is the most important greenhouse gas and is not photochemically active in the troposphere and stratosphere. Its concentration is controlled by burning of fossil fuel and by exchange with the ocean and biosphere. See discussion in Section 3.

N2O

Nitrous oxide, N_2O, is destroyed predominately in the stratosphere with a global mean lifetime of order 160 yr. Reductions in **upp-O3** lead to increased penetration of solar UV and to shorter lifetimes, and vice versa. Increases in stratospheric mixing rates (**circ**) lead to higher N_2O concentrations in the photodissociation region and hence to shorter lifetimes.

CH4

Methane, CH_4, is destroyed predominantly (~95 %) in the troposphere by reaction with trop-OH, and its lifetime should respond directly and immediately to changes in trop-OH with some sensitivity to tropospheric temperature (see later discussion of tropospheric chemistry). Stratospheric destruction, estimated to be less than 5% of the total, is treated as a constant, non-lagged loss in this model.

CFC13, CF2C12, C2F3C13, CC14, CF3Br, CF2ClBr, CF4

Perhalogenated hydrocarbons include: the chlorofluorocarbons, $CFCl_3$, CF_2Cl_2 and $C_2F_3Cl_3$; carbon tetrachloride, CCl_4; the bromofluorocarbons (halons), CF_3Br and CF_2ClBr; and carbon tetrafluoride, CF_4. Stratospheric loss represents the only major loss for these species, and thus their lifetimes are parameterized as functions of **upp-O3** and **circ**, as for N_2O. The lifetime for CF_4 is so long that loss over the next century is insignificant.

CH3CC13, CHF2Cl, CH3Cl, CH3Br

Hydrohalocarbons include: the solvent, CH_3CCl_3; the freon, CHF_2Cl; and the naturally occurring methyl halides, CH_3Cl and CH_3Br. In general these species' loss is dominated by reactions with tropospheric OH as for CH_4, and we calculate their loss as proportional to **trop-OH**. Lifetimes for the reference atmosphere (1985) are based on a scaling of the lifetime for CH_3CCl_3.

3. CLIMATE VARIABLES

CO2

Carbon dioxide, CO_2, is the single most important greenhouse gas in projections of climate change to the end of the 21st century. About 50% of the emissions of CO_2 from burning of fossil fuel are removed from the atmosphere each year, and uptake by the ocean is believed to represent the major loss. The marine sink for fossil fuel CO_2 is strongly dependent upon the temperature, chemistry, biology and effective vertical mixing of the near surface ocean. Oceanic uptake occurs primarily in the mixed layer and thermocline, and secondarily, through deep convection at high latitudes. Changes in ocean circulation should affect concomitantly the CO_2 and tropospheric temperatures, as discussed below.

Oceanic uptake is parameterized here as proportional to the difference between the current atmospheric CO_2 concentration and the preindustrial CO_2 concentration, assumed to be 285 ppm. The time constant for this system, C_C (in years), is currently about 40 years. The prognostic equation is:

$$\mathbf{CO2}(t+1) = \mathbf{CO2}(t) + S_{CO2}(t) - [\mathbf{CO2}(t) - 285]/C_C(t)$$

The time constant C_C of 40 years may appear reasonable in that it results in calculated uptake that is consistent with today's observations; however, the time constant is not a constant, and the return to pre-industrial levels of CO_2 would take much longer.

In the warmer world predicted under higher CO_2 concentrations of the next century, the proportion of annual fossil fuel CO_2 emissions entering the ocean is almost certain to be less than today, because the near-surface ocean of the future will likely be warmer, more stratified, and have higher total carbon burden. These effects are included in the formulation of C_C (i.e., the explicit dependence on **trop-T** and **CO2**) but values for these coefficients, other than zero, await results from more physically detailed models. The possibility of mode changes in ocean circulation, or changes in biological cycling of carbon in the ocean, cannot be predicted in this model, but may be included as an external forcing by making C_C proportional to a specified time-dependent ocean-mixing parameter, **ocean**.

The other net source / sink of potential significance for CO_2 is the terrestrial biosphere. Net fluxes to or from the terrestrial biosphere could be included in this model only as part of the overall CO_2 emissions that are specified for each scenario (S's in Table 1c).

net-F

The net greenhouse forcing of the lower atmosphere plus land and ocean, **net-F** (W/m^2), is derived instantaneously as a function of the greenhouse gases, the current tropospheric temperature, and the planetary albedo. Following the formulation of Dickinson (1986) we write

$$\mathbf{net\text{-}F} = \mathbf{Q} - \mathbf{B} \times \mathbf{trop\text{-}T} - 340 \times \mathbf{albedo} \quad (W/m^2)$$

where **Q** is the change in radiative forcing at the top of the troposphere due to changes in greenhouse gases (e.g., $2xCO_2$ gives Q = 4.3 W/m^2), **B** specifies the

increase in tropospheric temperature required to restore radiative equilibrium at the tropopause (e.g., the value of **B** ranges from 3 to 1 $W/m^2/°C$ for a global climate sensitivity ranging from 1.5 to 4.5 °C for $2xCO_2$), and **albedo** represents the percent change in planetary albedo. In this model we have assumed a sensitivity of 4 °C and hence B = 1.075 $W/m^2/°C$. Individual contributions to **Q** associated with the different gases are represented as a linear or second-order expansion about the *pre-industrial* state of the atmosphere and are derived from the 1-D radiative-convective model of Lacis et al. (1981).

The climate variables (**net-F**, **Q**, **trop-T**, **albedo**) are defined relative to a *pre-industrial* atmosphere, rather than the current 1985 *reference* atmosphere. The climate system appears to be now beginning to respond to perturbations to this quasi-steady-state condition that we denote by the pre-industrial atmosphere: CO_2 = 285 ppm, CH_4 = 0.7 ppm, N_2O = 280 ppb, CFCs = null, **trop-T** = 0 °C, **albedo** = 0 %). The initial or reference value of **Q** (1.55 W/m^2) and **net-F** will be different from zero, reflecting the fact that the steady-state warming associated with the change in atmospheric composition from pre-industrial times (~1.5 °C) has not yet been realized. In particular, we choose values of **trop-T** and **albedo** for the reference atmosphere: **trop-T**$_o$ = 0.5 °C, and **albedo**$_o$ = 0.0 %. Thus climate change would continue beyond the reference state even if concentrations of all trace gases remained fixed.

trop-T

Changes to the mean tropospheric temperature, **trop-T**, are coupled directly with the ocean surface temperature. This dependent variable is used in the model as a surrogate for climate change; it directly affects tropospheric chemistry through the temperature dependence of kinetic rates and abundance of water vapor, **trop-H2O**. The mean tropospheric temperature could also be used to calculate first-order climate feedbacks on the natural sources of some of the trace gases, but this is outside the scope of this model. Changes to the vertical and latitudinal distribution of tropospheric temperature are often predicted for climate perturbations; but their impact on tropospheric chemistry is unknown and is not included in this formulation.

The quantity **trop-T** is integrated over an annual time-step starting with the reference value **trop-T**$_o$ for t = 1985,

$$\textbf{trop-T}(t+1) = \textbf{trop-T}(t) + \textbf{net-F}(t)/C_T(t),$$

where $C_T(t)$ is the effective annual ocean heat capacity ($W/m^2/°C$). As in the case for CO_2 uptake (C_C), the quantity C_T might be expected to decrease as the earth warms because the ocean will become more stratified, reducing the effective transfer with the deep ocean. However, if temperatures increase then the volume of ocean involved in heat exchange with the atmosphere increases and the process becomes more efficient, i.e., C_T increases. At the very least, such an ocean heat uptake parameter should be used with great caution. (As with **CO2** we have included a parameterized dependence of C_T on **trop-T** and **ocean**, but currently have zeroed these coefficients.)

trop-H2O

The variable **trop-H2O** refers to the percent change in mean tropospheric water vapor abundance, adopted here as 6.2% per °C from the saturation vapor pressure

curve near 25°C. Tropospheric water vapor is assumed to respond instantaneously to changes in tropospheric temperature and to maintain a constant distribution of relative humidity. Ideally, one should be able to use results from general circulation-climate models to derive the change in specific humidity with increased greenhouse forcing. The feedback of tropospheric water vapor on **net-F** is included in the coefficient **B** above. Perturbations to water vapor are calculated relative to the reference state (**trop-T_o**), not the pre-industrial atmosphere (**trop-T** = 0 °C).

ocean

The ocean is an important reservoir of heat and CO_2. Ocean circulation could change as a result of global warming, but there is no basis for modeling changes in ocean circulation at this time. The variable **ocean** refers to the percent change in ocean mixing and is specified within this model as a function of time for each scenario (Table 1c). A positive value corresponds to more rapid exchange with the deep ocean and hence a greater capacity for uptake of CO_2 and heat (i.e., smaller values of C_C and C_T). A large negative value (-99%) corresponds to an ocean in which deep water formation has shut down.

albedo

The variable **albedo** (%) includes the impact of changing clouds, photochemical aerosols, and volcanic aerosols as atmospheric constituents that influence the planetary albedo. The aerosol optical depth of the non-volcanically perturbed stratosphere is ~ 0.005 and is dominated by sulfuric acid particles formed by oxidation of upwardly transported OCS and SO_2. This natural optical depth could change by a factor of 2 over the next century due to changes in atmospheric OCS and SO_2, but exact atmospheric budgets for these species are difficult to deduce. More importantly, the optical depth of stratospheric aerosols can increase transiently to values exceeding 0.1 following large volcanic eruptions such as Agung in 1963 and El Chichon in 1982. In the free troposphere oxidation of $(CH_3)_2S$ and SO_2 is a source of sulfuric acid particles which are efficient cloud condensation nuclei. Adding further uncertainty, it has recently been hypothesized that changes in surface emissions of these gases might control cloudiness in the remote marine troposphere.

Changes in cloud-cover or aerosols of a magnitude that would significantly alter the energy balance of the planet (±½%) would have a similar impact (±½%) on the solar UV radiation in the troposphere, and these latter effects are not considered here. The major impact of **albedo** will be on the radiative forcing, **net-F**, as noted in the equation above. Due to the difficulty of explicitly modeling these changes, the current model assumes that **albedo** is given by an external scenario (Table 1c) and may include the impacts of changes in sea ice, snow cover and biospheric activity.

4. THE STRATOSPHERE

Stratospheric ozone controls the photochemical destruction of many atmospheric gases. Ozone is the primary source of chemically reactive species in the atmosphere, it competes for solar ultraviolet radiation that destroys many long-lived gases, and it also has a direct radiative effect on stratospheric temperatures. The effect on surface temperature is expected to be small for the expected change in ozone distribution. Treatment of perturbations to stratospheric ozone in this assessment model does not replace studies of stratospheric chemistry or of ozone's impact on ultraviolet radiation reaching the surface. Rather, this approach is used to determine changes in the stratospheric loss rates of long-lived gases and in the tropospheric chemistry that is controlled by the stratospheric ozone column (i.e., OH).

col-O3 and **upp-O3**

Stratospheric ozone is represented by two variables: **col-O3** refers to the total stratospheric plus tropospheric column; and **upp-O3** refers to ozone in the upper stratosphere, the column above 30 km. The selection of these two ozone variables is based on the different processes that control them, as well as the different impacts they have on the lifetimes of the long-lived gases.

The variable **upp-O3** represents the percent change in the column of O_3 above 30 km with respect to the reference atmosphere (circa 1985). Current predictions of ozone change (WMO, 1986) show significant decline in upper stratospheric ozone as compared with the pre-CFC atmosphere (circa 1950). In the upper stratosphere, ozone responds directly and rapidly to changing levels of odd-nitrogen (NO_y), chlorine (Cl_x), water vapor and methane, as well as changes in stratospheric temperature (perturbed primarily by CO_2). For example, the response of ozone above 30 km to increased Cl_x is fairly linear, but increases in CH_4 can partly offset increases in Cl_x. The NO_y and Cl_x catalytic cycles do not interfere so much with each other in the upper stratosphere. We account for the necessary first and second order dependencies, and some cross-terms. These sensitivities have been derived from one-dimensional chemical models in the current assessment model.

Decreases in **upp-O3** result in increased levels of ultraviolet radiation throughout the middle and lower stratosphere, and hence, in increased destruction rates for many of the long-lived gases. A further impact is on the total ozone column: reduced mixing ratios of O_3 in the upper stratosphere mean that less ozone is transported into the lower stratosphere, but this reduction is cancelled in large part by the increased production of odd-oxygen (O_3+O) caused by the enhanced UV levels lower in the atmosphere.

The variable **col-O3** refers to the percent change in globally integrated ozone relative to the reference atmosphere. The total column of ozone includes contributions from both the upper stratosphere (~10 %) and the troposphere (~10 %), but resides predominantly in the lower stratosphere. Predicting ozone perturbations in the lower stratosphere is more complicated than in the upper stratosphere because the ozone abundance is controlled by transport as well as chemistry and the existence of UV radiative feedbacks from the changes in ozone in the column above (**upp-O3**). Photochemical loss of ozone is dominated by NO_y reactions in the middle and lower stratosphere, but Clx interferes with the NOy

catalytic cycle here. The primary impact of Br_x on ozone is predicted to occur in the lower stratosphere and is coupled to Cl_x levels. Breakdown of methane affects ozone differently in the lower and upper stratosphere: in the lower stratosphere HO_2 released from CH_4 oxidation reacts with NO to produce more ozone, while in the upper stratosphere the increased levels of HO_2 (from either CH_4 or H_2O) destroy O_3 directly. The importance of transport (**circ**) in controlling **col-O3** is clear from the long photochemical lifetimes of O_3 in the lower stratosphere, but the magnitude of the perturbation associated with a change in the circulation has not yet been evaluated.

Changes in concentrations of trace gases could change stratospheric temperatures due to the direct radiative effects of the trace gases or indirectly through their effect on O_3. A change in temperature will in turn modulate the effectiveness of odd-oxygen destruction mechanisms, resulting in a change in O_3. These effects have been included directly in the sensitivity factors relating ozone to the particular trace gases. For example, **col-O3** and **upp-O3** are parameterized as a function of **CO2** in spite of the fact that CO_2 has no direct chemical reactions in the stratosphere, because of the predicted change in stratospheric temperatures.

The purpose of predicting column ozone is to calculate tropospheric changes in the solar ultraviolet radiation, which is the primary source of OH. Further, change in **col-O3** that occurs predominantly in the lower stratosphere may have a direct radiative effect on climate, and can control a major source of tropospheric O_3.

We have limited the perturbation to the total ozone column, **col-O3**, to ±20 % (1) because of uncertainties in our understanding of how the atmosphere would respond to large ozone decreases and (2) to avoid treatment of higher order nonlinear feedbacks. Similarly, Cl_x levels are limited to 20 ppb or less to avoid these highly nonlinear interactions. The upper stratosphere is driven more directly by Cl_x, and **upp-O3** values as large as ±60 % are allowed.

str-Clx and **str-Brx**

Total inorganic chlorine and bromine, **str-Clx** and **str-Brx**, refers to the Cl_x mixing ratio (ppb) near 40 km and the Br_x mixing ratio (ppt) near 25 km (again, averaged over latitude and season). We assume that **str-Clx** and **str-Brx** are equal to the sum of all chlorine and bromine atoms, respectively, contained in the source gases listed in Table 1a. The sources include all chlorinated and brominated hydrocarbons (with a time lag of 3 years), and the coefficients reflect the number of Cl and Br atoms released by each gas near 40 and 25 km, respectively.

The distribution of Cl_x and Br_x within the stratosphere varies with altitude and latitude and depends on the rate of photochemical destruction of the various source gases. The present model uses single values for **str-Clx** and **str-Brx** to parameterize the effect on ozone: the major impact of Clx on ozone is in the upper stratosphere above 30 km (**upp-O3**), whereas the major impact of Br_x on ozone occurs below 30 km (**col-O3**). Stratospheric levels of Cl_x and Br_x may also be affected by the strength of the circulation through the fraction of Cl and Br atoms released from the source gases, but this effect is probably small and not included in the present formulation.

str-NOy

Predicting the concentration of total stratospheric odd nitrogen, NO_y, is qualitatively more difficult than for Cl_x and Br_x because NO_y is produced in only a few percent of the reactions destroying its predominant source gas, N_2O. Furthermore, the reactions between NO_y species produce N_2, providing a chemical sink for NO_y within the stratosphere, and thus conservation of odd-nitrogen cannot be invoked. In the model, **str-NOy** represents the maximum average mixing ratio of about 18 ppb occurring in the tropics between 30-40 km in the present day atmosphere.

Changes in **str-NOy** are based on increases or decreases in abundance of the source gas N_2O, applying a time lag of 2.5 years to the mean tropospheric concentration **N2O** as discussed above for the long-lived gases. Perturbations to other known (but lesser) sources of stratospheric NO_y (tropospheric lightning, thermospheric and mesospheric NO, ionization by cosmic rays) are not included in this model. Stratospheric losses for NO_y vary from quadratic in NO_y concentrations (i.e., involving reaction of two odd-nitrogen species, N and NO) to linear (i.e., transport in the troposphere and removal to the surface). Therefore, a negative term proportional to the square of **N2O** is included to simulate the effect of increased, non-linear, chemical removal at elevated levels of NO_y.

The distribution of NO_y in the lower stratosphere is also sensitive to the circulation which controls the rate of removal from the stratosphere and the advective transport of upper tropospheric NO_y into the lower tropical stratosphere. The present approach of representing the effect of NO_y by a single parameter precludes such detailed treatments. It should be kept in mind that the present treatment assumes that there is no significant change in the vertical structure of NO_y in the perturbed state so that the effect on both **col-O3** and **upp-O3** can be adequately parameterized in terms of **str-NOy**.

str-H2O

The abundance of stratospheric water is believed to be controlled by the concentration of water vapor at the tropical tropopause (**str-H2O**) and the abundance of CH_4. The model does not currently predict changes in the amount of water entering the tropical stratosphere, although changing climate is expected to influence stratospheric water through modification of the tropopause temperature and tropical convection or, possibly, through polar stratospheric clouds. The reference mixing ratio for **str-H2O** of 3.0 ppm can be altered by external specification of the scenario.

circ

The rate of circulation in the stratosphere is defined by the average vertical flux of air in the tropics (±30° latitude) across the tropopause and through the lower stratosphere, and is denoted by **circ**. The sign convention adopted is that this quantity is positive (+%) when the circulation and exchange is more rapid than in the reference state.

A faster circulation will shorten the lifetimes of the source gases by transporting greater concentrations of the long-lived gases into the upper stratosphere where they are more readily destroyed. The impact can be estimated with current 1-D and 2-D models by changing the rate of vertical diffusion (1-D) or the magnitude of the diabatic circulation (2-D). These effects are coupled nicely through the

1½-D conceptual models (Mahlman et al, 1986; Holton, 1986). The circulation can also affect the lifetimes indirectly by changing the distributions of other photochemically active trace species such as Cl_x, NO_y, Br_x and O_3, but this effect is probably secondary. A faster circulation may also lead to modest redistribution of column O_3 from the tropics to the midlatitudes. The net impact on tropospheric OH (higher in the tropics and lower at midlatitudes) is not estimated here and may cancel at least partially on a global scale.

Changes in **circ** are most likely driven by alterations to the wave forcing of the stratosphere by the troposphere (i.e., climate or more specifically **trop-T**) or by local radiative heating (i.e., **CO2**, **str-H2O**, **col-O3** and **upp-O3**), but cannot be predicted currently. Thus we leave **circ** as an externally forced variable (similar to **ocean**) that couples parametrically to the lifetimes of the long-lived gases.

5. TROPOSPHERIC CHEMISTRY

Oxidation of many atmospheric species is initiated primarily by reactions with the hydroxyl radical OH, and secondarily by reactions with ozone. The oxidizing capacity of the troposphere determines the abundance of many gases such as methane, carbon monoxide, methyl chloroform and CHF_2Cl. We focus therefore on predicting perturbations to the global mean levels of OH and O_3 in the troposphere. The prediction of trends in global tropospheric models is at present a difficult research problem, complicated especially by our lack of knowledge of the global distribution of NO_x. Thus our predictions of changes in tropospheric ozone must necessarily be assigned large uncertainties.

The simulation of the troposphere in this model includes separate treatment of the two hemispheres (NH & SH) because significant asymmetries are observed in many of the important shorter-lived gases such as CO, NO_x and non-methane hydrocarbons. These species play a major role in the budgets for O_3 and OH in each hemisphere. Many of these species, especially OH, are highly variable, and averaging even over a hemisphere may not adequately represent their interactions with other trace gases.

trop-OH

The variable **trop-OH** refers to the percent perturbation with respect to the reference state of the global, annual mean value for tropospheric OH concentrations. **Trop-OH** is a combination of the independently calculated northern hemisphere (**NH-OH**) and southern hemisphere (**SH-OH**) values that we assume to be equally weighted for the reference state.

$$\textbf{trop-OH} = 0.50 \times \textbf{NH-OH} + 0.50 \times \textbf{SH-OH}$$

In global averaging the OH concentrations are weighted by the kinetic reaction rates and the density of air. For a well mixed gas such as CH_4 the average loss frequency would include a weighting kernel containing density and kinetic rates,

$$\langle OH \rangle_{CH_4} = \langle OH_{local} \times M \times \exp(-1700/T) \rangle \; / \; \langle M \times \exp(-1700/T) \rangle,$$

where **M** is the background air density and the averaging, < >, is performed over each hemisphere. This formula applies to the global troposphere and ideally to multi-dimensional models for tropospheric OH. In view of the fact that global models for these type of calculations are not available, we can only note that the average values **trop-OH**, **NH-OH** and **SH-OH** should reflect this weighting kernel.

The value **trop-OH** is treated as a steady-state variable; it is assumed to respond immediately to the annual average values of the trace gases. To derive perturbations to OH we solve a non-linear system, equating a "production" term to a "loss" term. For each hemisphere of the reference atmosphere (**NH-OH** = **SH-OH** ~ 0 %), we define the partitioning of OH loss among the predicted gases (CH_4, CO), the specified fluxes (NMHC), and self-reactions (OH). The production side of the equation includes a positive response to increased UV (i.e., loss in column ozone) and to increases in tropospheric H_2O, O_3 and NO_x fluxes. Coefficients for variations in either the production or loss terms with respect to column O_3 (**col-O3**), tropospheric water vapor (**trop-H2O**), trop-O_3 (**NH/SH-O3**), and CO (**NH/SH-CO**), CH_4 (**CH4**), and fluxes of both NMHCs and NO_x are based on results from 1-

D and 2-D models (Liu et al., 1987; Thompson & Cicerone, 1986; Isaksen & Hov, 1987). Major sources of uncertainty in calculating OH are the spatial averaging for this highly variable constituent and the nonlinearity in perturbation coefficients, especially with respect to NO_x flux (see Figure 1, Liu et al., 1987)

The sensitivity, $d\ln\langle OH\rangle/d\ln[S_{NOx}]$, of average tropospheric OH to emissions of NO_x is predicted to be positive for levels of NO_x ranging from a few ppt up to 1-10 ppb and to be negative at NO_x levels above about 1-10 ppb. This effect will be significant in the mid-latitude continental boundary layer (below 3 km). Since global <OH> is dominated by clean air for current levels of NO_x (1-10 ppt NO_x over the oceans; 20-200 ppt over the continents), the OH versus NO_x response curves are currently in the regime where an increase of NO_x by a factor of 2, yields OH increases of only 25 % in the boundary layer. The impact on global <OH> is likely to be less than 10 % for a doubling of anthropogenic NO_x emissions from industrial and agricultural regions. The dependence of OH on CH_4 is straightforward, but seasonality of CO and NMHCs are important and must be incorporated into models from which an annual average is derived. Some of the sensitivities used here are based on the average of OH over four seasons, including the observed seasonality of CO (e.g., Logan et al., 1981).

The first-order dependence of **NH-OH and SH-OH** on tropospheric temperature, **trop-T**, has been included. Increasing temperatures will increase the kinetic rate coefficients for OH reactions with gases such as CH_4 and CH_3CCl_3 by about 2% per °C. If these gases were the only sink for tropospheric OH, then concentrations of OH would decrease by a similar amount, cancelling the effect of a rise in tropospheric temperature. The reaction of OH with CO has no temperature dependence and represents approximately half of the loss of tropospheric OH. Thus, an increase in **trop-T** alone would shift relative loss fractions, increasing loss fractions of CH_4 and decreasing loss fractions of CO.

NH-O3 & SH-O3

Perturbations to tropospheric ozone, **NH-O3** and **SH-O3** (% change), will affect both tropospheric temperatures and the long-lived source gases controlled by OH. A significant fraction of tropospheric ozone originates in the stratosphere and is destroyed by surface deposition; it is sufficiently short-lived (a few months) that we must calculate ozone perturbations separately for each hemisphere. In the simplified model adopted here, we assume that sources (stratospheric ozone and tropospheric "smog" chemistry) respond to atmospheric composition and that loss frequencies (photochemical and surface reactions) remain constant.

Changes in the stratospheric source of tropospheric ozone are associated with perturbations to the total ozone column, **col-O3**, since these perturbations are reflected primarily in the mixing ratio of O_3 in the lower stratosphere. Tropospheric chemical reactions also contribute significantly to the ozone budget. For ozone concentrations in the NH, sensitivity coefficients, $d\ln(O_3)/d\ln(X)$, can be ascribed to the precursor gases (CH_4, 0.2; CO, 0.1; NOx flux, 0.1; NMHC flux, 0.1) based on detailed photochemical models for typical tropospheric air parcels (Liu et al., 1987; Thompson et al., 1988) The combined effect of changes in tropospheric composition is assumed to be linear, but uncertainties in the coefficients are large, approximately a factor of 2. Unfortunately, the efficiency of O_3 production varies widely with the NO_x levels (Liu et al., 1987) which in turn cannot be adequately characterized throughout the entire troposphere due to their large

dynamic range. In the SH the efficiency for production of ozone by these precursors is similar to that in the NH; however, variability is less.

Photochemical loss within the troposphere occurs primarily through reactions of $O(^1D)$ with H_2O and of HO_2 with O_3, and would, to first-order, become more efficient as tropospheric water increases. In regions with elevated concentrations of NO_x and NMHCs, however, an increase in H_2O and hence HO_2 may enhance production of O_3 (Liu et al., 1987). Because of this ambiguity a specific dependence on **trop-H2O** and **trop-T** has not been included. In some remote areas with low levels of NO_x an increase in highly reactive NMHC emissions (e.g., isoprene) may lead to increased near-surface destruction of ozone. In order to construct a better model for global tropospheric ozone, it is necessary to have a realistic model that incorporates the wide distribution of NO_x concentrations and that includes the impact of the large range of NMHCs.

Observations show an increase in NH continental ozone which probably is in the range of 10-60% over the last 100 years. In the SH there are insufficient observations to form similar conclusions. As observations are limited to continental areas, the trends over oceans (and thus the global trends) are not known. Increases in **NH-O3** over the last two decades seem to be in the range of a few tenths to one percent per year, but the uncertainty in these numbers may be as large as ±1 %/yr.

NH-CO & SH-CO

Observations of CO have reported various northern hemispheric trends in this gas over the past decade. While the evidence indicates that CO is increasing at these sites on the order of 1±1 %/yr, it is not clear whether this is a year-round or hemispheric change. Increasing concentrations of CO can lead directly to a decrease in OH concentrations and, more indirectly, to increased production of tropospheric O_3.

Carbon monoxide has a hemispherically averaged lifetime of about 3 months, dominated by loss in the tropics. Within each hemisphere the gas is moderately well mixed when compared with NO_x and NMHCs, but still exhibits large, factor-of-two, variations with season and adjacent to sources. We choose to define and calculate an annual mean concentration of CO within each hemisphere: **NH-CO** and **SH-CO** (ppb). Seasonal variations of CO are observed and are critical for accurate modeling of tropospheric OH and for averaging over the annual cycle. Sources include methane oxidation (proportional to OH), NMHC oxidation (proportional to annual flux), and direct emissions of CO (biosphere, combustion). Loss is proportion to OH in each hemisphere, and interhemispheric transport from NH to SH is included through the use of a single transfer coefficient assuming an exchange residence time of 1 yr.

NH/NOx & SH/NOx

NO_x is the rate-limiting precursor for photochemical production of O_3 throughout most of the troposphere, except in regions dominated by anthropogenic emissions of NO_x and hydrocarbons. NO_x also plays a key role in determining the distribution of OH. The concentrations of NO_x vary over three orders of magnitude throughout the troposphere. The lifetime of NO_x against photochemical conversion to HNO_3 or loss on surfaces varies from hours to weeks, and sources are patchy. Thus, the global distribution of NO_x is expected to be extremely

heterogeneous. A "typical" concentration cannot be easily defined, and therefore, we choose to define perturbations to mean tropospheric NO_x "levels" as being linearly proportional to the *flux* of NO_x in each hemisphere.

The effective *flux* of NO_x (Tg(N)/yr of $NO+NO_2$) into the troposphere of each hemisphere is denoted by **NH/NOx** and **SH/NOx**, and is specified in the scenario (S's in Table 1c). The current global sources of NO_x include stratospheric HNO_3 (~1 TgN/yr), lightning (8 TgN/yr), biogenic emissions (8 TgN/yr), combustion (both surface and aircraft, 21 TgN/yr), and biomass burning (12 TgN/yr). Most of the NO_x emissions are in the NH. For the SH we assume that the effective sources are lightning (3 TgN/yr) biogenic emissions (3 TgN/yr), combustion (1 TgN/yr), and biomass burning (6 TgN/yr). (Note, in the sample calculation the fluxes **NH/NOx** and **SH/NOx** were assumed to be 20 and 10 TgN/yr respectively.)

There is a large temporal and spatial variation in these emissions. Furthermore these sources are not simply additive: most of the NO released from the surface (combustion or biospheric sources) is oxidized and removed before becoming mixed into the more homogeneous "free troposphere." Production of O_3 associated with this NO_x therefore depends on the local photochemical environment, particularly the abundance of hydrocarbons. Surface emissions are also subject to rapid non-linear losses in highly polluted regions and are likely to have less impact (on a per molecule basis) than NO_x sources distributed throughout the troposphere, such as lightning and aircraft. These issues are unresolved in the current models of global tropospheric chemistry and predicting the impact of NO_x on O_3 is highly uncertain. Without more detailed global models for tropospheric NO_x, we have chosen to treat these sources as linearly additive, but recognize the potential for error in this assumption.

NH/NMHC & SH/NMHC

Non-methane hydrocarbons (NMHCs) also play an important role in the production of tropospheric O_3 and in the levels of OH. The problem of describing NMHCs is similar to that for NO_x in that a "typical" concentration cannot be easily defined for each hemisphere. Furthermore, the title NMHC includes all hydrocarbons other than methane: C_2H_2, C_2H_6, C_3H_8, and isoprene (C_5H_8) among others. Each species has different concentrations and different reactivities with OH and sometimes O_3. Lifetimes of the longest lived species such as C_2H_6 are typically a few months. Thus, all NMHCs are effectively removed within a year, and we assume (as for NO_x) that their impact is proportional to their annual *flux*. Their oxidation in the troposphere generally consumes OH and produces CO and O_3.

The *flux* of NMHCs (Tg(C)/yr) into the troposphere of each hemisphere is denoted by **NH/NMHC** and **SH/NMHC**, and is specified in the scenario (S's in Table 1c). The base level of NMHCs, 0.0%, corresponds to a natural background source of 1000 Tg(C) per year in the northern hemisphere, and 500 Tg(C) per year in the southern hemisphere. In comparison, the anthropogenic source is only about 70 Tg(C) per year. The problems associated with intense boundary-layer sources and the inhomogeneous distribution of tropospheric NO_x applies also to NMHCs.

6. RESULTS OF MODEL CALCULATIONS

A sample scenario for the emission of trace gases has been chosen to demonstrate the assessment model. The scenario, defined in Table 1e, describes the rate of change in emissions (%/yr) from the reference atmosphere (1985) into the 21st century. The initial fluxes are given as Pg/yr (for ppm units), Tg/yr (for ppb units), Gg/yr (for ppt units). For those gases with large and uncertain natural sources, the scenario specifies the currently observed rate of growth, and the model calculates a consistent starting flux using the initial lifetime and observed trend.

This particular emissions scenario is one of several supplied by the EPA and represents a modest to low-growth scenario for combustion with a Montreal Protocol limitation on CFCs. Fluxes of N_2O, CH_4 and CO_2 increase by an average of about 0.5, 0.8 and 1.2 %/yr, respectively, over the next five decades. Fluxes of $CFCl_3$ and CF_2Cl_2 decease slightly after 1990; whereas CHF_2Cl emissions increase by about 5 %/yr over the next 30 years. After a small decline, CH_3CCl_3 is predicted to have sustained growth of 2.5 %/yr after 1990. The halons exhibit rapid growth (~10 %/yr) until 2000 with a reduced but sustained growth (0.3 - 6 %/yr) thereafter. The combustion sources of CO are predicted to decrease through the next century; and anthropogenic emissions of NO_x increase (~0.5 %/yr) after 2010. This report makes no attempt to justify the details of the EPA scenarios, but we chose one exhibiting growth as an example of the simulation of a rapidly changing atmosphere.

Coefficients from six parameters were chosen to represent the uncertainty in the model predictions:

(1) the lifetime for (oceanic) uptake of CO_2 was varied from 40 yr to 60 yr;
(2) the feedback of tropospheric temperature on **net-F** (i.e., **B**) was varied from 0.75 to 1.25, corresponding to a feedback factor of 2.5 to 4.5;
(3) the current lifetime of $CFCl_3$ was varied from 55 yr to 75 yr;
(4) the non-linear response of total column ozone to stratospheric chlorine was increased by a factor of 7, from -0.03 to -0.20 %/ppb^2;
(5) the sensitivity of tropospheric O_3 to CH_4 abundance in the northern hemisphere was varied from 0.20 to 0.40; and
(6) the sensitivity of northern-hemispheric OH production to NO_x emissions was varied from 0.05 to 0.20.

The model results are shown in Figures F1-F8. The set of calculations from each case (2^6 = 64 total) gives a mean value from the 64 trace-gas histories plus an upper and lower range represented by ±2 times the rms variance about the mean time line. Interestingly, in this scenario the model calculations stopped in 2038 because total stratospheric bromine exceeded the specified upper limit of 100 ppt. For some gases such as CH_4 or CO, these uncertainties have generated a significant spread in the predicted concentrations by 2035, but for the gases dominated by stratospheric loss, no uncertainty range is demonstrated by this particular example. Clearly more effort must be made to understand how well the model represents this uncertainty.

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7. CONCLUSIONS: A CRITICAL REVIEW

The following statements are taken verbatim from the replies of the participants and reviewers.

Overall

"This is an idealized heuristic model whose primary function should be regarded as providing the first step toward a more realistic prediction of future changes in atmospheric composition. Its strengths lie in the important recognition that atmospheric constituents have specific sensitivities to and feedbacks involving both other constituents and climate. Its weaknesses revolve around its inability to include many of these sensitivities and feedbacks in a physically realistic way."

"A simple model for atmospheric composition can only be as good as the more complex models on which it is based."

"I am still distressed by the willful naivete of the hyper-simple model. It seems to me that we need to be very careful about our assessments of the uncertainty limits for various processes. We may have to be particularly cautious about those [parameters] in which the fundamental processes are either poorly understood or the nonlinearity is extreme, such as tropospheric NO_x and O_3."

"The model attempts to cover all the important processes and feedbacks in a sound manner, and the nature of the approximations and uncertainties inherent in such a model are indicated. Commenting primarily on the tropospheric results, I note that they are consistent with those of other models in predicting changes in CO-OH-O_3, etc. on the 50-100 year time frame."

"I believe that [this model] has done an excellent job in setting out the various couplings and clearly stating the caveats that must be observed. In particular I have no problem with your treatment of the stratospheric component."

Caveats

"The ozone and temperature changes provided by the model are only for the purpose of adjusting the lifetimes of the trace gases, and not to be used independently for assessment purposes."

"The current parameterized model is not suitable for direct assessment of the impact of trace gases on climate or stratospheric ozone, but may be valuable for internal use by the EPA and others to differentiate between emission scenarios."

"[The] general philosophy is a good one, provided the model can be used as an interpolation device to provide simple numerical summaries corresponding to known behavior of the more elaborate, physically realistic models. For this to work the model needs a) checks to insure it is not used outside its range of validity and b) a process to update it to current understanding every year or two."

Uncertainties and Scenarios

"The number of cases used to encompass the range of values for key coefficients should be about $2^{7\pm1}$, and resulting trace gas histories must show both the mean concentration and ±2 standard deviations."

"The choice of scenarios for emissions of trace gases is one of the more important elements in applications of this model; predicting the growth in emissions (outside the scope of this workshop) represents perhaps the greatest uncertainty in predicting future composition."

"The main difficulties with predicting changes in tropospheric composition are that (1) present-day budgets of key source gases are uncertain so that we don't know what 'baseline emissions' are and (2) the projections of CO, NO etc. emissions vary greatly. Thus the limitations in this model output may come primarily from the assumed scenarios more than approximations made in model chemistry and physics."

"As such, uncertainties in emission scenarios dominate over uncertainties in the model (already noted ...), it is not clear what this model gains over nothingness."

"Despite these doubts, I guess that I feel that it is better public policy to go ahead and use a model such as this one to make preliminary assessments rather than simply giving up and doing nothing because we can't represent all the couplings and feedbacks. To do nothing would indicate that the level of uncertainty was higher than it probably is."

Work still to do

"The one area where I would like to see a further stressing of the uncertainties is the albedo-cloudiness feedback problem. This seems to me to be absolutely crucial for the climate problem. Yet I sense that surprisingly little serious work is being done in this area of a fundamental nature."

"With regards the current formulation, I have one significant concern: that is, I'm uncomfortable with the way you do atmospheric CO_2 with only a single ocean reservoir; I believe you need at least 2 such boxes. As currently formulated, CO_2 goes to preindustrial values in less than 100 years, whereas the correct time scale is several thousand. The 40-year relaxation time that you use applies only to the ocean above the main thermocline, i.e., the first 600m, or so. If ... all emissions [were to cease] now, the ocean would decrease atmospheric CO_2 little in the next 40 years, perhaps by 10-20 ppm but not 65 ppm."

"I am concerned that the report suggests that current modelling capabilities for tropospheric OH and ozone are more advanced than is in fact the case. The report discusses the non-linear dependence of OH and ozone on NO_x; [but] the assessment model does not account for this non-linearity. Instead, fluxes of NO_x are averaged hemispherically, a procedure which gives concentrations for NO_x which are always in the linear regime. Current understanding of the dependence of ozone on NO_x and hydrocarbons suggests that the averaging inherent in one and two dimensional models prevents them from giving reliable predictions of trends in ozone, since they cannot treat separately regions at the same latitude

with high and low NO_x. The assessment model has the same problem, since it is based on 1-d and 2-d models."

"The treatment of hydrocarbons is also a difficult area. The effect of hydrocarbons on OH and ozone depends on the amount of NO_x, and the assessment model does not account for this. The question of the effect of changes in NO_x and hydrocarbons on global ozone and OH has not been examined sufficiently with research models for there to be adequate information to derive a reliable assessment model."

"This parameterization depends much on the current understanding of the physics and chemistry of the atmosphere and the atmospheric budgets of trace gases. These are likely to change in the future. There shouid be a built-in structure and mechanism for reviewing the inputs in the model."

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TABLE 2. Program Listing

```
C---(COMMBLK.)-----Atmospheric Composition Model--COMMON BLOCKS---
      IMPLICIT REAL*4(A-H,O-Z), INTEGER*4(I,J,K,M,N), LOGICAL*4(L)
      CHARACTER*10 YTIT,YCOD,XTIT,XCOD,ZTIT,ZCOD
      CHARACTER*30 YCTIT,YCTITL,XCTIT,XCTITL,TBLANK,TBLOCK, FTIT
      CHARACTER*80 TITLE,TITLE2,TITLE3
C---
      COMMON/TITLES/ YTIT(16),XTIT(15),ZTIT(12),YCOD(16),XCOD(15),
     X   YCTIT(18,16),XCTIT(18,15), FTIT(8),
     X   YCTITL,XCTITL, TITLE,TITLE2,TITLE3
      COMMON/VARXY/ YATWT(16),XATWT(15), YATMF(16),XATMF(15),
     X  YSCAL(16),XSCAL(15), YC(18,16),XC(18,15),
     X  YT(122,16),XT(122,15), YTP(122,16),ZTP(122,12), YTL(122,16)
C---
      COMMON/XSOLV/ X(15),FX(15),AX(15,15),DELX(15),MXSLV
      COMMON/NNNNNN/ N,NDX,NY0,NY1,NMAX,NPRTH, MX,MY,MZ,KMAX
      COMMON/LLLLLL/ LSTOP,LPRTI, NOUT
      COMMON/SSSSSS/ PHIY0(16),PHIZ0(12), PHIDEC(12),
     X  PHIYT(12,16),PHIZT(12,12), KY0(16)
C---
      COMMON/FUZZ/ FYT1(122,16),FYT2(122,16),FXT1(122,15),FXT2(122,15),
     X             FXYC0(8),FXYC1(8),KFX(8),KFY(8),KFC(8),NFZZ
C---
      DATA NXSET,MYSET,MXSET,KXSET /122, 16, 15, 18/
      DATA TBLANK/'                                '/
      DATA TBLOCK/' AT-COMP commons: v3.0   08/88'/

C---(ATCOMP.for)-------Atmospheric Composition Model------version 3.   (08/88)
C----------------based on Workshop at NASA/GISS, NYC, January 1988-----------
C---reads UNIT=1, reference list from DATAREF., specific to the current coding
C---reads UNIT=2, parameters from DATA., titles must match those in DATA.
C---reads UNIT=3, flux data from FLUX.
C---writes UNIT=6, default output in 80 column format
C---writes UNIT=7, data file for spreadsheets, post-processing (N.B. 2**N runs)
C---writes UNIT=8, data file for post-processing, means +- 2 std dev's
C---
C---compiled with MS FORTRAN V-4.10, requires 80x87 support, example of run:
C---      C>atcomp dataref data flux unit7out unit8out > output
C---running with 2**6=64 cases, shutdown in 2038 (53 yrs), takes 2 min on
C---         a COMPAQ 386/20, a full up scenario (128 x 115 yrs) = about 9 min.
C---
$INCLUDE:'commblk.'
C--------
      CALL READIN
        N = NY0-1
        NDX = 0
        NDXMIN = NY1-NY0+1
      CALL INIT
C---calculate fluxes for all years
      CALL FLUXES
C---do manifold of solutions with pairs of coefficients:
      NFZDO = 2**NFZZ
      DO 20 II=1,NFZDO
C---binary decompose to select high (FXYC1) or low (FXYC0) coeff's
        IIF = II-1
        WRITE(6,*) ' RUN #',II
        DO 10 JJ=NFZZ,1,-1
           JJ2 = 2**(JJ-1)
           IIJ = IIF/JJ2
          KX = KFX(JJ)
          KY = KFY(JJ)
          KC = KFC(JJ)
        IF(IIJ.EQ.1) THEN
          IF(KX.GT.0) XC(KC,KX) = FXYC1(JJ)
          IF(KY.GT.0) YC(KC,KY) = FXYC1(JJ)
C         WRITE(NOUT,101) ' ----high value:',FTIT(JJ),FXYC1(JJ),KX,KY,KC
        ELSE
          IF(KX.GT.0) XC(KC,KX) = FXYC0(JJ)
```

TABLE 2 (continued)

```
          IF(KY.GT.0) YC(KC,KY) = FXYC0(JJ)
C         WRITE(NOUT,101) ' ----low  value:',FTIT(JJ),FXYC0(JJ),KX,KY,KC
        ENDIF
   10   IIF = IIF - JJ2*IIJ
C---
C--------Begin on Jan 1, NY0 and integrate through to Jan 1, NY1
C---check for limits
        LSTOP = .FALSE.
      DO 11 NN=NY0,NY1
         N = NN-1
         NDX = NN-NY0
C---calculate steady-state species during year = N
        IF(NDX.GT.0) CALL SSYEAR
C---integrate long-lived species to beginning of year = N+1
        IF(NDX.GT.0) CALL LLYEAR
C---check that limits are not exceeded
        CALL LIMITS
         IF(LSTOP) GOTO 12
   11 CONTINUE
        N = NY1-1
   12 CONTINUE
        NDXMIN = MIN0(NDX,NDXMIN)
      CALL PRTALL(NY0,N+1)
C---ACCUMULATE MEANS/VARIANCES
      DO 15 I=1,MY
      DO 14 NX=1,NDX+1
      FYT1(NX,I) = FYT1(NX,I) + YT(NX,I)
   14 FYT2(NX,I) = FYT2(NX,I) + YT(NX,I)**2
   15 CONTINUE
      DO 18 I=1,MX
      DO 17 NX=1,NDX
      FXT1(NX,I) = FXT1(NX,I) + XT(NX,I)
   17 FXT2(NX,I) = FXT2(NX,I) + XT(NX,I)**2
   18 CONTINUE
C---
   20 CONTINUE
C---PRINT OUT SUMMARY:
      DO 22 I=1,MY
      DO 21 NX=1,NDXMIN+1
        XBAR = FYT1(NX,I)/FLOAT(NFZDO)
        XRMS = FYT2(NX,I)/FLOAT(NFZDO) - XBAR*XBAR
        XRMS = SQRT(AMAX1(0.0,XRMS))
      YT(NX,I)   = XBAR
      FYT1(NX,I) = XBAR - 2.*XRMS
   21 FYT2(NX,I) = XBAR + 2.*XRMS
   22 CONTINUE
      DO 26 I=1,MX
      DO 25 NX=1,NDXMIN
        XBAR = FXT1(NX,I)/FLOAT(NFZDO)
        XRMS = FXT2(NX,I)/FLOAT(NFZDO) - XBAR*XBAR
        XRMS = SQRT(AMAX1(0.0,XRMS))
      XT(NX,I)   = XBAR
      FXT1(NX,I) = XBAR - 2.*XRMS
   25 FXT2(NX,I) = XBAR + 2.*XRMS
   26 CONTINUE
C---file output for post-processing:
        NY1M = MIN0(NY1,NY0+NDXMIN)
      WRITE(8,102) TITLE3
      WRITE(8,103) (YTIT(I), I=1,MY)
      DO 31 NN=NY0,NY1M,NPRTH
       NX = NN-NY0+1
       WRITE(8,104) NN,(YT(NX,I), I=1,MY)
   31 CONTINUE
      WRITE(8,105) (YTIT(I), I=1,MY)
      DO 32 NN=NY0,NY1M,NPRTH
       NX = NN-NY0+1
       WRITE(8,104) NN,(FYT1(NX,I), I=1,MY)
   32 CONTINUE
      WRITE(8,106) (YTIT(I), I=1,MY)
      DO 33 NN=NY0,NY1M,NPRTH
```

TABLE 2 (continued)

```
        NX = NN-NY0+1
        WRITE(8,104) NN,(FYT2(NX,I), I=1,MY)
   33 CONTINUE
      WRITE(8,103) (XTIT(I), I=1,MX)
      DO 35 NN=NY0,NY1M-1,NPRTH
        NX = NN-NY0+1
        WRITE(8,104) NN,(XT(NX,I), I=1,MX)
   35 CONTINUE
      WRITE(8,105) (XTIT(I), I=1,MX)
      DO 36 NN=NY0,NY1M-1,NPRTH
        NX = NN-NY0+1
        WRITE(8,104) NN,(FXT1(NX,I), I=1,MX)
   36 CONTINUE
      WRITE(8,106) (XTIT(I), I=1,MX)
      DO 37 NN=NY0,NY1M-1,NPRTH
        NX = NN-NY0+1
        WRITE(8,104) NN,(FXT2(NX,I), I=1,MX)
   37 CONTINUE
C---
  101 FORMAT(1X,A20,A20,1P,E10.3,3I5)
  102 FORMAT(A80)
  103 FORMAT('year',30(',',A7))
  104 FORMAT(I4,30(',',F7.2))
  105 FORMAT(' MIN',30(',',A7))
  106 FORMAT(' MAX',30(',',A7))
      STOP
      END

      SUBROUTINE READIN
C---read in all parameters
$INCLUDE:'commblk.'
C---basic setups:
      KMAX = KXSET
      NMAX = NXSET
      LSTOP = .FALSE.
      LPRTI = .FALSE.
      NOUT = 6
        NY0 = 1980
        NY1 = 2100
        NPRTH = 1
      WRITE(NOUT,100) TBLOCK
C---read programmed list of parameters from UNIT=1:
      READ(1,100) TITLE
        WRITE(NOUT,100) TITLE
      READ(1,102) MY
        WRITE(NOUT,205) MY,MYSET
        IF(MY.GT.MYSET) STOP
      READ(1,100)
      DO 12 II=1,MY
       READ(1,102)  I
       DO 10 K=1,KMAX
       READ(1,101) YCTIT(K,I)
       IF(YCTIT(K,I).EQ.TBLANK) GOTO 11
   10  CONTINUE
   11  CONTINUE
   12 CONTINUE
      READ(1,102) MX
        WRITE(NOUT,206) MX,MXSET
        IF(MX.GT.MXSET) STOP
      READ(1,100)
      DO 16 II=1,MX
       READ(1,102)  I
       DO 14 K=1,KMAX
       READ(1,101) XCTIT(K,I)
       IF(XCTIT(K,I).EQ.TBLANK) GOTO 15
   14  CONTINUE
   15  CONTINUE
   16 CONTINUE
      CLOSE (1)
C---read in parameters from UNIT=2, check against programmed list
```

TABLE 2 (continued)

```
      READ(2,100) TITLE2
        WRITE(NOUT,100) TITLE2
      READ(2,100)
      READ(2,100)
      DO 24 II=1,MY
        READ(2,102)  I,YCOD(I),YTIT(I),YATWT(I),YATMF(I),YSCAL(I)
        WRITE(NOUT,201) I,YTIT(I),YCOD(I),YATWT(I),YATMF(I),YSCAL(I)
          DO 20 K=1,KMAX
   20     YC(K,I) = 0.0
        DO 21 K=1,KMAX
         READ(2,103) YC(K,I), YCTITL
         IF(YCTITL.NE.YCTIT(K,I)) THEN
           WRITE(NOUT,202) YTIT(I),I,K,YC(K,I),YCTITL,YCTIT(K,I)
           STOP
         ENDIF
         IF(YCTITL.EQ.TBLANK) GOTO 22
         IF(LPRTI) WRITE(NOUT,203) YCTITL, YC(K,I)
   21   CONTINUE
   22   CONTINUE
   24 CONTINUE
      READ(2,100)
      READ(2,100)
      DO 28 II=1,MX
        READ(2,102)  I,XCOD(I),XTIT(I),XATWT(I),XATMF(I),XSCAL(I)
        WRITE(NOUT,204) I,XTIT(I),XCOD(I),XATWT(I),XATMF(I),XSCAL(I)
          DO 25 K=1,KMAX
   25     XC(K,I) = 0.0
        DO 26 K=1,KMAX
         READ(2,103) XC(K,I), XCTITL
         IF(XCTITL.NE.XCTIT(K,I)) THEN
           WRITE(NOUT,202) XTIT(I),I,K,XC(K,I),XCTITL,XCTIT(K,I)
           STOP
         ENDIF
         IF(XCTITL.EQ.TBLANK) GOTO 27
         IF(LPRTI) WRITE(NOUT,203) XCTITL, XC(K,I)
   26   CONTINUE
   27   CONTINUE
   28 CONTINUE
      CLOSE (2)
C---read in flux data from UNIT=3
      READ(3,100) TITLE3
        WRITE(NOUT,100) TITLE3
      READ(3,105) NY0,NY1,NPRTH
        NY1 = MIN0(NY1,NY0+NMAX-1)
      READ(3,100) TITLE
        WRITE(NOUT,100) TITLE
      DO 31 I=1,MY
      DO 30 J=1,12
   30 PHIYT(J,I) = 0.0
   31 PHIY0(I) = 0.0
      DO 33 II=1,MY
C---currently, FLUX = flux in Xg/yr,   PHIY0(I) = flux in Xg/yr
C---           PHIYT(DECADE,I) = growth rate (%/yr) during a DECADE: 1 = 1980s
C---           KY0(I) = 0 (true flux), = 1 (growth rate, calculate PHIY0)
        READ(3,104)  I,YCTITL,K,FLUX,PHIDEC
        IF(I.LT.1 .OR. I.GT.MY) GOTO 34
        PHIY0(I) = FLUX
        KY0(I) = K
        DO 32 J=1,12
   32   PHIYT(J,I) = PHIDEC(J)
        WRITE(NOUT,207) I,YCTITL,K,FLUX,PHIDEC
   33 CONTINUE
   34 CONTINUE
C---setups for external scenarios: albedo, ocean, circ, CO, NOx, NMHCs  (PHIZ-)
C---MUST BE IN ORDER
      READ(3,100)
      DO 36 I=1,12
      DO 35 J=1,12
   35 PHIZT(J,I) = 0.0
      ZTIT(I) = TBLANK
```

TABLE 2 (continued)

```
   36 PHIZO(I) = 0.0
      DO 38 II=1,12
        READ(3,104)  I,ZCOD,K,FLUX,PHIDEC
        IF(I.LT.1) GOTO 39
        ZTIT(I) = ZCOD
        PHIZO(I) = FLUX
        MZ = I
        DO 37 J=1,12
   37   PHIZT(J,I) = PHIDEC(J)
        WRITE(NOUT,207) I,ZCOD,K,FLUX,PHIDEC
   38 CONTINUE
   39 CONTINUE
C---read in uncertainty ranges for coeff's from UNIT=3, pick either YC or XC
      DO 48 I=1,8
      READ(3,105) KKFY,KKFX,KKFC
      IF(KKFC.LT.1) GOTO 49
      IF(KKFY.GT.0) KKFX = 0
      READ(3,103) FXYC0(I), FTIT(I)
      READ(3,103) FXYC1(I), FTIT(I)
         IF(KKFY.GT.0 .AND. FTIT(I).NE.YCTIT(KKFC,KKFY)) THEN
           WRITE(NOUT,202)
     W         YTIT(KKFY),KKFY,KKFC,FXYC1(I),FTIT(I),YCTIT(KKFC,KKFY)
           STOP
         ENDIF
         IF(KKFX.GT.0 .AND. FTIT(I).NE.XCTIT(KKFC,KKFX)) THEN
           WRITE(NOUT,202)
     W         XTIT(KKFX),KKFX,KKFC,FXYC1(I),FTIT(I),YCTIT(KKFC,KKFX)
           STOP
         ENDIF
      KFY(I) = KKFY
      KFX(I) = KKFX
      KFC(I) = KKFC
   48 CONTINUE
      I = 9
   49 NFZZ = I-1
      CLOSE (3)
      RETURN
  100 FORMAT(A80)
  101 FORMAT(10X,A30)
  102 FORMAT(I2,1X,A7,A10,5E10.3)
  103 FORMAT(E10.3,A30)
  104 FORMAT(I2,A8,I1,E9.2,12F5.2)
  105 FORMAT(16I5)
  201 FORMAT(' Y-species=',I3,1X,A10,A7,' atwt:atmf:Tg/ppb=',3F10.3)
  202 FORMAT('  ERROR:  MISMATCH ON INPUT: ',A10,2I4,1PE10.3,2A30)
  203 FORMAT(1X,A30,1PE12.3)
  204 FORMAT(' X-species=',I3,1X,A10,A7,' atwt:atmf:Tg/ppb=',3F10.3)
  205 FORMAT('  no. Y-species/dim:',2I10)
  206 FORMAT('  no. X-species/dim:',2I10)
  207 FORMAT(1X,I2,A8,I2,1P,E10.3,0P,12F6.2)
      END

      SUBROUTINE INIT
C---initialize all species for year=Jan 1, NYR0(=N), called with NDX=0
$INCLUDE:'commblk.'
C---zero all arrays:
      DO 3 I=1,MY
       DO 2 NN=1,NMAX
       FYT1(NN,I) = 0.0
       FYT2(NN,I) = 0.0
       YT(NN,I) =  0.0
       YTP(NN,I) = 0.0
    2  YTL(NN,I) = 0.0
    3 CONTINUE
      DO 5 I=1,MX
       DO 4 NN=1,NMAX
       FXT1(NN,I) = 0.0
       FXT2(NN,I) = 0.0
    4  XT(NN,I) = 0.0
    5 CONTINUE
```

TABLE 2 (continued)

```
      DO 7 I=1,MZ
       DO 6 NN=1,NMAX
    6  ZTP(NN,I) = 0.0
    7 CONTINUE
C---initialize concentrations:
      DO 12 I=1,MY
   12 YT(1,I) = YC(1,I)
      DO 14 I=1,MX
   14 XT(1,I) = XC(1,I)
C---initialize fluxes to get current growth rate:  PHIY0() in %/yr
      DO 22 I=1,MY
      IF(KY0(I).LT.1) GOTO 22
       YLOSS = YT(1,I)/YC(4,I)
       IF(I.EQ.3) YLOSS = YLOSS - YC(5,I)/YC(4,I)
       YPROD = YSCAL(I)*( YLOSS + 0.01*PHIY0(I)*YT(1,I) )
       PHIY0(I) = YPROD
   22 CONTINUE
      RETURN
      END

      SUBROUTINE LIMITS
C---check for exceeding limits:
$INCLUDE:'commblk.'
C---check for all Y-species for year=N+1  (end of year=N)
      DO 10 I=1,MY
      IF (YT(NDX+1,I).LT.YC(2,I) .OR. YT(NDX+1,I).GT.YC(3,I)) THEN
        WRITE(NOUT,101) N,YTIT(I),YCOD(I),YT(NDX+1,I),YC(2,I),YC(3,I)
        LSTOP = .TRUE.
      ENDIF
   10 CONTINUE
C---check for all X-species for year=N    (average over year=N)
      IF(NDX.LE.0) GOTO 99
      DO 20 I=1,MX
      IF (XT(NDX,I).LT.XC(2,I) .OR. XT(NDX,I).GT.XC(3,I)) THEN
        WRITE(NOUT,101) N,XTIT(I),XCOD(I), XT(NDX,I),XC(2,I),XC(3,I)
        LSTOP = .TRUE.
      ENDIF
   20 CONTINUE
   99 RETURN
  101 FORMAT(' >>>>>LIMITS>>>>',I4,' FOR ',A10,1X,A7,' VALUE=',F8.2,
     X       '  LIMITS=',2F8.2)
      END

      SUBROUTINE FLUXES
C---calculate fluxes (Gg/yr) for years NY0 (Jan 1 - Dec 31) thru year NY1
C---    for all species (long-lived & external scenarios & trop chem)
$INCLUDE:'commblk.'
C---fluxes for long-lived species, initial rates:
      DO 12 I=1,MY
   12 YTP(1,I) = PHIY0(I)
C---fluxes for external scenarios:  albedo, ocean, circ, NH/SH-NOx, CO, NMHC
      DO 14 I=1,MZ
   14 ZTP(1,I) = PHIZ0(I)
C---growth in fluxes each year:
      DO 28 NN=NY0+1,NY1-1
        NNDX = NN+1-NY0
        NDEC = MIN0(12,MAX0(1, (NN/10) - 197 ))
      DO 22 I=1,MY
   22 YTP(NNDX,I) = YTP(NNDX-1,I)*(1.0 + 0.01*PHIYT(NDEC,I))
      DO 24 I=1,MZ
   24 ZTP(NNDX,I) = ZTP(NNDX-1,I)*(1.0 + 0.01*PHIZT(NDEC,I))
   28 CONTINUE
      RETURN
      END

      SUBROUTINE SSYEAR
C---calculate steady-state species for the year Jan 1-Dec 31 of year=N
C---    uses long-lived species for Jan 1 and fluxes from the whole year
$INCLUDE:'commblk.'
C---certain X-species depend only on the explicitly integrated
```

TABLE 2 (continued)

```
C---     long-lived species and are calculated explicitly each year
C---     from the long-lived species on Jan 1.
C---
C---a set of X-species depend on each other and are solved implicitly
C---     each year as a set of coupled non-linear equations.
C---these species must occupy the first MXSLV elements of the XT vector
C
C---XT(NDX,I) contains a first guess (last-year's value) for the constituents
      IF(NDX.GE.1) THEN
        DO 2 I=1,MX
    2   XT(NDX,I) = XT(NDX-1,I)
      ENDIF
C---perts to ref atmos:  N2O, CH4, CO2, trop-T
      YT1 = YT(NDX,1)-YC(1,1)
      YT2 = YT(NDX,2)-YC(1,2)
      YT3 = YT(NDX,3)-YC(1,3)
      YT15= YT(NDX,15)-YC(1,15)
C---[11]:  str-NOy    (use lagged/last-year's upp-O3 for feedback)
      XT(NDX,11) = XC(1,11) + XC(4,11)*YT1 + XC(5,11)*YT1*YT1
     X + XC(6,11)*XT(NDX,10)*0.01
C---[12]:  str-Clx
      XT(NDX,12) =
     X + XC(4,12)*YT(NDX,4) + XC(5,12)*YT(NDX,5) + XC(6,12)*YT(NDX,6)
     X + XC(7,12)*YT(NDX,7) + XC(8,12)*YT(NDX,8) + XC(9,12)*YT(NDX,9)
     X + XC(10,12)*YT(NDX,10)
C---[13]:  str-Brx
      XT(NDX,13) =
     X   XC(4,13)*YT(NDX,11) + XC(5,13)*YT(NDX,12) + XC(6,13)*YT(NDX,13)
C---[14]:  str-H2O
      XT(NDX,14) = XC(1,14) + XC(4,14)*YT15
C---perts to ref atmos:  str-NOy, Clx, Brx, H2O
      XT11 = XT(NDX,11)-XC(1,11)
      XT12 = XT(NDX,12)-XC(1,12)
      XT13 = XT(NDX,13)-XC(1,13)
      XT14 = XT(NDX,14)-XC(1,14)
C---[10]:  upp-O3   (can now do explicitly here)
      XT(NDX,10) = XC(4,10)*XT11 + XC(5,10)*XT12 + XC(6,10)*YT2
     X  + XC(7,10)*YT2*XT12 + XC(8,10)*XT14 + XC(9,10)*YT3
C---[ 9]:  col-O3   (can now do explicitly here)
      XT(NDX,9) = XC(4,9)*XT(NDX,10) + XC(5,9)*XT11 + XC(6,9)*XT12
     X + XC(7,9)*XT13 + XC(8,9)*YT2 + XC(9,9)*XT14 + XC(10,9)*ZTP(NDX,3)
     X + XC(11,9)*XT11*XT12 + XC(12,9)*XT12*XT13 + XC(13,9)*XT12*YT2
     X + XC(14,9)*XT12*XT12 + XC(15,9)*YT3
C---[ 8]: trop-H2O   wrt ref atmos
      XT(NDX,8) = 100.*( (1.0 + XC(4,8))**YT15 - 1.0 )
C---
C---      begin iteration for coupled non-linear system:
C---X(I) is the value of XT(NDX,I) on each successive iteration
C---first guess is filled in the XT(NDX,I) array
      DO 10 I=1,MX
   10 X(I) = XT(NDX,I)
C---inplicitly solved for quantities:  XT(NDX,1-7)
      MXSLV = 7
      CALL NEWRAF
C---have solved for coupled system, now finish explicitly derived quantities:
      DO 20 I=1,MXSLV
   20 XT(NDX,I) = X(I)
C---[15]: net-F     wrt pre-industrial atmos (285->300 ppm = 0.257 W/m2)
      XT(NDX,15) = XC(1,15)
     X  - XC(4,15) * YT15            - XC(5,15)*ZTP(NDX,1)
     X  + XC(6,15)*(YT(NDX,3)-300.) + XC(7,15)*(YT(NDX,3)-300.)**2 +.257
     X  + XC(8,15)*(YT(NDX,2)-700.) + XC(9,15)*(YT(NDX,1)-280.)
     X  + XC(10,15)*YT(NDX,4)       + XC(11,15)*YT(NDX,5)
     X  + XC(12,15)*YT(NDX,6)       + XC(13,15)*YT(NDX,7)
     X  + XC(14,15)*0.5*(XT(NDX,2)+XT(NDX,5))
   99 RETURN
      END

      SUBROUTINE SSEVAL
C---calculate Jacobian and right-hand-side for the steady-state species = X(I)
```

TABLE 2 (continued)

```
C---     solve FX(i) = 0,     del(FX(i))/del(X(j)) = AX(i,j)
$INCLUDE:'commblk.'
C---clear the AX-matrix
      DO 12 J=1,MXSLV
       FX(J) = 0.0
        DO 11 I=1,MXSLV
   11   AX(I,J) = 0.0
   12 AX(J,J) = 1.0
C---have solved all the strat-quantities explicitly, do coupled troposphere
C---  alone, collapse to 7x7 system
C---perts to ref atmos:  trop-T
      YT15= YT(NDX,15)-YC(1,15)
C---[1]:   trop-OH
      FX(1) = X(1) - XC(4,1)*X(3) - XC(5,1)*X(6)
      AX(1,1) = 1.0
      AX(1,3) = -XC(4,1)
      AX(1,6) = -XC(5,1)
C---[2]:   NH-O3
      FX(2) = XC(4,2)*XT(NDX,9) + 100.*( XC(5,2)*(YT(NDX,2)/YC(1,2)-1.)
     X + XC(6,2)*(X(4)/XC(1,4)-1.) + XC(7,2)*(ZTP(NDX,6)/ZTP(1,6)-1.)
     X + XC(8,2)*(ZTP(NDX,8)/ZTP(1,8)-1.) )   -  X(2)
      AX(2,2) = -1.0
      AX(2,4) = 100.*XC(6,2)/XC(1,4)
C---[3]:   NH-OH ( % )      FX = P/Po - L/Lo   (Po = Lo)  ***include ref offset
C---             P = J(O'D)*O3*H2O + NOx*(HO2 + ROO)
C---             L = OH*( CH4 + CO + NMHCs + HOx)
       X3 = 1. + .01*(X(3) + XC(1,3))
      XPROD = 1. + .01*( XC(4,3)*XT(NDX,8) + XC(5,3)*X(2)
     P + XC(6,3)*XT(NDX,9) )  + XC(7,3)*(ZTP(NDX,6)/ZTP(1,6) - 1.)
      XLOSS = (1.-XC(8,3)-XC(9,3)-XC(10,3)-XC(11,3))
     L + XC(8,3)*(1.+YC(6,2)*YT15)*YT(NDX,2)/YC(1,2)
     L + XC(9,3)*X(4)/XC(1,4) + XC(10,3)*ZTP(NDX,8)/ZTP(1,8)
     L + XC(11,3)*X3
      FX(3) = XPROD - X3*XLOSS
      AX(3,2) = +.01*XC(5,3)
      AX(3,3) = -.01*(XLOSS + XC(11,3)*X3)
      AX(3,4) = -X3*XC(9,3)/XC(1,4)
C---[4]:   NH-CO (ppb)      0 = P(CH4) + P(NMHC) + P(CO-flux) - L(N-S) - L(OH)
        XLOSS = (1. + .01*X(3)*XC(8,4))/XC(4,4)
        YPROD = XC(5,4)*(YT(NDX,2)/YC(1,2))*(1.0 + YC(6,2)*YT15)
      FX(4) = YPROD*(1.+.01*X(3))
     X + XC(6,4)*ZTP(NDX,8)/ZTP(1,8) + ZTP(NDX,4)/XSCAL(4)
     X - (X(4)-X(7))/XC(9,4) - X(4)*XLOSS
      AX(4,3) = .01*YPROD - X(4)*.01*XC(8,4)/XC(4,4)
      AX(4,4) = -1./XC(9,4) - XLOSS
      AX(4,7) = +1./XC(9,4)
C---[5]:   SH-O3 ( % )
      FX(5) = XC(4,5)*XT(NDX,9) + 100.*( XC(5,5)*(YT(NDX,2)/YC(1,2)-1.)
     X + XC(6,5)*(X(7)/XC(1,7)-1.) + XC(7,5)*(ZTP(NDX,7)/ZTP(1,7)-1.)
     X + XC(8,5)*(ZTP(NDX,9)/ZTP(1,9)-1.) )   -  X(5)
      AX(5,5) = -1.0
      AX(5,7) = 100.*XC(6,5)/XC(1,7)
C---[6]:   SH-OH ( % )      FX = P/Po - L/Lo   (Po = Lo)
       X6 = 1. + .01*(X(6) + XC(1,6))
      XPROD = 1. + .01*( XC(4,6)*XT(NDX,8) + XC(5,6)*X(5)
     P + XC(6,6)*XT(NDX,9) )  + XC(7,6)*(ZTP(NDX,7)/ZTP(1,7) - 1.)
      XLOSS = (1.-XC(8,6)-XC(9,6)-XC(10,6)-XC(11,6))
     L + XC(8,6)*(1.+YC(6,2)*YT15)*YT(NDX,2)/YC(1,2)
     L + XC(9,6)*X(7)/XC(1,7) + XC(10,6)*ZTP(NDX,9)/ZTP(1,9)
     L + XC(11,6)*X6
      FX(6) = XPROD - X6*XLOSS
      AX(6,5) = +.01*XC(5,6)
      AX(6,6) = -.01*(XLOSS + XC(11,6)*X6)
      AX(6,7) = -X6*XC(9,6)/XC(1,7)
C---[7]:   SH-CO (ppb)      0 = P(CH4) + P(NMHC) + P(CO-flux) - L(S-N) - L(OH)
        XLOSS = (1. + .01*X(6)*XC(8,7))/XC(4,7)
        YPROD = XC(5,7)*(YT(NDX,2)/YC(1,2))*(1.0 + YC(6,2)*YT15)
      FX(7) = YPROD*(1.+.01*X(6))
     X + XC(6,7)*ZTP(NDX,9)/ZTP(1,9) + ZTP(NDX,5)/XSCAL(7)
     X - (X(7)-X(4))/XC(9,7) - X(7)*XLOSS
```

TABLE 2 (continued)

```
      AX(7,6) = .01*YPR00 - X(7)*.01*XC(8,7)/XC(4,7)
      AX(7,7) = -1./XC(9,7) - XLOSS
      AX(7,4) = +1./XC(9,7)
      RETURN
      END

      SUBROUTINE LLYEAR
C---calculate long-lived species for Jan 1, year=N begin Jan 1, year=N-1
C---   uses steady-state species averaged over year=N
$INCLUDE:'commblk.'
C---calculate losses (YTL's) for the year:
      CALL LLLOSS
C---simple annual integrator:
      DO 20 I=1,MY
      YT(NDX+1,I) = YT(NDX,I) + (YTP(NDX,I) - YTL(NDX,I))/YSCAL(I)
   20 CONTINUE
      RETURN
      END

      SUBROUTINE LLLOSS
C---calculate annual loss for long-lived species = YTL (Xg/yr)
C---   for year beginning Jan 1, N (NDX), includes strat-lag
C---hardwired to parameterization given in UNIT=1
$INCLUDE:'commblk.'
C---XT1 = % pert to trop OH (avg of NH + SH)
C---XT10 = % perturb to upp-O3
C---ZT2 = % pert to ocean (circ) (+ = faster)
C---ZT3 = % pert to circ (strat) (+ = faster)
C---YT3 = degrees pert of CO2 wrt to ref atmos
C---YT15= degrees pert to trop T wrt to ref atmos
      XT1 = 0.01*XT(NDX,1)
      XT9 = 0.01*XT(NDX,9)
      XT10= 0.01*XT(NDX,10)
      ZT2 = 0.01*ZTP(NDX,2)
      ZT3 = 0.01*ZTP(NDX,3)
      YT3 = YT(NDX,3)-YC(1,3)
      YT15= YT(NDX,15)-YC(1,15)
      NDXLAG = MAXO(NDX-2,1)
C---1=N2O
      YLIFE = YC(4,1) * (1.0 + YC(5,1)*XT10 + YC(6,1)*ZT3)
      YTL(NDX,1) = YT(NDXLAG,1)*YSCAL(1)/YLIFE
C---2=CH4
      YLIFE = YC(4,2) * (1.0 + YC(5,2)*XT1 - YC(6,2)*YT15)
      YTL(NDX,2) = YT(NDX,2)*YSCAL(2)/YLIFE
C---3=CO2
      YLIFE = YC(4,3) * (1.0 + YC(6,3)*ZT2 + YC(7,3)*YT15 + YC(8,3)*YT3)
      YTL(NDX,3) = (YT(NDX,3) - YC(5,3))*YSCAL(3)/YLIFE
C---4=CFCl3
      YLIFE = YC(4,4) * (1.0 + YC(5,4)*XT10 + YC(6,4)*ZT3)
      YTL(NDX,4) = YT(NDXLAG,4)*YSCAL(4)/YLIFE
C---5=CF2Cl2
      YLIFE = YC(4,5) * (1.0 + YC(5,5)*XT10 + YC(6,5)*ZT3)
      YTL(NDX,5) = YT(NDXLAG,5)*YSCAL(5)/YLIFE
C---6=CHFCl2
      YLIFE = YC(4,6) * (1.0 + YC(5,6)*XT1 - YC(6,6)*YT15)
      YTL(NDX,6) = YT(NDX,6)*YSCAL(6)/YLIFE
C---7=C2F3Cl3
      YLIFE = YC(4,7) * (1.0 + YC(5,7)*XT10 + YC(6,7)*ZT3)
      YTL(NDX,7) = YT(NDXLAG,7)*YSCAL(7)/YLIFE
C---8=CCl4
      YLIFE = YC(4,8) * (1.0 + YC(5,8)*XT10 + YC(6,8)*ZT3)
      YTL(NDX,8) = YT(NDXLAG,8)*YSCAL(8)/YLIFE
C---9=CH3CCl3
      YLIFE = YC(4,9) * (1.0 + YC(5,9)*XT1 - YC(6,9)*YT15)
      YTL(NDX,9) = YT(NDX,9)*YSCAL(9)/YLIFE
C---10=CH3Cl
      YLIFE = YC(4,10) * (1.0 + YC(5,10)*XT1 - YC(6,10)*YT15)
      YTL(NDX,10) = YT(NDX,10)*YSCAL(10)/YLIFE
C---11=CF3Br
      YLIFE = YC(4,11) * (1.0 + YC(5,11)*XT10 + YC(6,11)*ZT3)
```

TABLE 2 (continued)

```
      YTL(NDX,11) = YT(NDXLAG,11)*YSCAL(11)/YLIFE
C---12=CF2BrCl
      YLIFE = YC(4,12) * (1.0 + YC(5,12)*XT9 + YC(6,12)*ZT3)
      YTL(NDX,12) = YT(NDXLAG,12)*YSCAL(12)/YLIFE
C---13=CH3Br
      rLIFE = YC(4,13) * (1.0 + YC(5,13)*XT1 - YC(6,13)*YT15)
      YTL(NDX,13) = YT(NDX,13)*YSCAL(13)/YLIFE
C---14=CF4
      YLIFE = YC(4,14)
      YTL(NDX,14) = YT(NDXLAG,14)*YSCAL(14)/YLIFE
C---15=trop-T  define forcing = net-F/lifetime, no loss
      YLIFE = YC(4,15) * (1.0 + YC(5,15)*ZT2 + YC(6,15)*YT15)
      YTP(NDX,15) = XT(NDX,15)/YLIFE
      RETURN
      END

      SUBROUTINE PRTALL(N1,N2)
C---prints & stores results of time-dependent integration from year=N1 thru N2
$INCLUDE:'commblk.'
C---Y-time series of long-lived species
        IYDO = (MY+9)/10
      DO 28 IY=1,IYDO
        I1 = 10*(IY-1)+1
        I2 = MIN0(I1+9,MY)
       WRITE(NOUT,101)
       WRITE(NOUT,106) (YTIT(I), I=I1,I2)
       WRITE(NOUT,102) (YCOD(I), I=I1,I2)
       DO 20 NN=N1,N2,NPRTH
         NX = NN+1-NY0
         WRITE(NOUT,103) NN, (YT(NX,I), I=I1,I2)
   20  CONTINUE
       IF(NPRTH.GT.1) GOTO 28
       WRITE(NOUT,104)
C      WRITE(NOUT,102) (YCOD(I), I=I1,I2)
       DO 22 NN=N1,N2-1,NPRTH
         NX = NN+1-NY0
         WRITE(NOUT,103) NN, (YTP(NX,I), I=I1,I2)
   22  CONTINUE
       WRITE(NOUT,105)
C      WRITE(NOUT,102) (YCOD(I), I=I1,I2)
       DO 24 NN=N1,N2-1,NPRTH
         NX = NN+1-NY0
         WRITE(NOUT,103) NN, (YTL(NX,I), I=I1,I2)
   24  CONTINUE
   28 CONTINUE
C---X-time series of steady-state species
        IXDO = (MX+9)/10
      DO 38 IX=1,IXDO
        I1 = 10*(IX-1)+1
        I2 = MIN0(I1+9,MX)
       WRITE(NOUT,107)
       WRITE(NOUT,106) (XTIT(I), I=I1,I2)
       WRITE(NOUT,102) (XCOD(I), I=I1,I2)
       DO 30 NN=N1,N2-1,NPRTH
         NX = NN+1-NY0
         WRITE(NOUT,103) NN, (XT(NX,I), I=I1,I2)
   30  CONTINUE
   38 CONTINUE
C---file output for post-processing:
        WRITE(7,203) TITLE3
        WRITE(7,201) (YTIT(I), I=1,MY)
      DO 42 NN=N1,N2,NPRTH
        NX = NN+1-NY0
        WRITE(7,202) NN,(YT(NX,I), I=1,MY)
   42 CONTINUE
        WRITE(7,201) (XTIT(I), I=1,MX)
      DO 46 NN=N1,N2-1,NPRTH
        NX = NN+1-NY0
        WRITE(7,202) NN,(XT(NX,I), I=1,MX)
   46 CONTINUE
```

TABLE 2 (continued)

```
C---
  101 FORMAT(/'  MIXING RATIOS/TEMPERATURE:')
  102 FORMAT(' year ',10A7)
  103 FORMAT(1X,I4,10F7.1)
  104 FORMAT(/'  PROD (ppt=Gg/yr, ppb=Tg/yr, ppm=Pg/yr)')
  105 FORMAT(/'  LOSS (ppt=Gg/yr, ppb=Tg/yr, ppm=Pg/yr)')
  106 FORMAT('      ',10(1X,A6))
  107 FORMAT(/'  PERTURBATIONS (steady-state):')
  201 FORMAT('year',30(',',A7))
  202 FORMAT(I4,30(',',F7.2))
  203 FORMAT(A80)
      RETURN
      END

      SUBROUTINE NEWRAF
C----drive program for NEWTON-RAPHSON non-linear system solver:
C----   uses subroutine SSEVAL to evaluate function (FX) and Jacobian (AX)
C----   uses subroutines LINSLV & RESOLV to solve the matrix equation
$INCLUDE:'commblk.'
C--------
      FERR = 1.0E-8
      XERR = 2.0E-5
C----   BEGIN NEWTON-RAPHSON:
      DO 20 ITER=1,30
           CALL SSEVAL
      DO 12 I=1,MXSLV
   12 FX(I) = -FX(I)
           CALL LINSLV(AX,FX,DELX,MXSLV,15)
           CALL RESOLV(AX,FX,DELX,MXSLV,15)
      DO 16 I=1,MXSLV
   16 X(I) = X(I) + DELX(I)
C     WRITE(6,'(4H DX=,1P,8E9.2)') (DELX(I), I=1,MXSLV)
C----   CHECK FOR CONVERGENCE:
      FMAX = 0.0
      XMAX = 0.0
      DO 18 I=1,MXSLV
      FMAX = AMAX1(ABS(FX(I)),FMAX)
      XMAX = AMAX1( ABS(DELX(I))/(ABS(X(I))+1.E-10), XMAX)
   18 CONTINUE
C---  IF(FMAX.LT.FERR) GOTO 22
      IF(XMAX.LT.XERR) GOTO 22
   20 CONTINUE
C----   END NEWTON-RAPHSON:
      WRITE(6,*) '  *******STEADY-STATE FAILED TO CONVERGE, YEAR:',N
      WRITE(6,'(4H FX=,1P,8E9.2)') (FX(I), I=1,MXSLV)
      WRITE(6,'(4H  X=,1P,8E9.2)') (X(I), I=1,MXSLV)
      WRITE(6,'(4H DX=,1P,8E9.2)') (DELX(I), I=1,MXSLV)
      GOTO 99
   22 CONTINUE
C          CALL SSEVAL
C     WRITE(6,*) '  -------HAVE CONVERGED------'
C     WRITE(6,'(4H  X=,1P,10E9.2)') (X(I), I=1,MXSLV)
C     DO 32 I=1,MXSLV
C     WRITE(6,'(4H  J=,1P,10E9.2)') (AX(I,J), J=1,MXSLV)
C  32 CONTINUE
   99 RETURN
      END

           SUBROUTINE LINSLV(A,B,X,N,NDIM)
C----- SUB -LINSLV- SOLVES MATRIX EQUATION:  A(N,N)*X(N) = B(N)
C-----    BY REDUCING THE A-MATRIX IN PLACE, WITH PARTIAL PIVOTING.
C----- SUB -RESOLV- ASSUMES THAT THE A-MATRIX HAS BEEN PROPERLY REDUCED
C-----     AND JUST SOLVES FOR X(N).  THIS OPTION ALLOWS THE SYSTEM
C-----     TO BE RESOLVED WITH A NEW B-VECTOR.
      REAL*4 A(NDIM,NDIM),B(NDIM),X(NDIM)
      REAL*8 S,SMAX,DIV,SUM
      INTEGER*4 N,NDIM,I,IPA,J,JP,K,KR,KRMAX
      COMMON/PASS/ S(30),IPA(30)
C-----
      DO 20 KR=1,N
```

TABLE 2 (continued)

```
      DO 11 K=1,N
   11 S(K) = A(K,KR)
        IF(KR.EQ.1) GOTO 14
      DO 12 J=1,KR-1
        JP = IPA(J)
        A(J,KR) = S(JP)
        S(JP) = S(J)
        DO 12 I=J+1,N
   12 S(I) = S(I) - A(I,J)*A(J,KR)
   14 KRMAX = KR
      SMAX = DABS(S(KR))
      DO 15 I=KR,N
        IF(DABS(S(I)).LT.SMAX) GOTO 15
        KRMAX = I
        SMAX =  DABS(S(I))
   15 CONTINUE
      IPA(KR) = KRMAX
      A(KR,KR) = S(KRMAX)
      DIV = 1.0D0/S(KRMAX)
      S(KRMAX) = S(KR)
      DO 16 I=KR+1,N
   16 A(I,KR) = S(I)*DIV
   20 CONTINUE
      RETURN
      END
      SUBROUTINE RESOLV(A,B,X,N,NDIM)
C  BACK SOLUTION FOLOWS REDUCTION BY -LINSLV- FOR GIVEN VALUE OF B()
      REAL*4 A(NDIM,NDIM),B(NDIM),X(NDIM)
      REAL*8 S,SMAX,DIV,SUM
      INTEGER*4 N,NDIM,I,IP,IPA,J
      COMMON/PASS/ S(30),IPA(30)
C-----
      DO 21 I=1,N
   21 S(I) = B(I)
      DO 22 I=1,N
        IP = IPA(I)
        X(I) = S(IP)
        S(IP) = S(I)
        DO 22 J=I+1,N
   22   S(J) = S(J) - A(J,I)*X(I)
      DO 25 I=N,1,-1
        SUM = X(I)
        DO 24 J=I+1,N
   24   SUM = SUM - A(I,J)*X(J)
   25 X(I) = SUM/A(I,I)
      RETURN
      END
```

TABLE 3. Model Coefficients

```
 UNIT=1    DATA REFERENCE STD (8/88) VERSION-3.
15 long-lived species = Y's
 # code    title          at.wt. atm.frac.    Tg/ppb
01 A(ppb) N2O(N)           28.02     0.96      4.78 nitrous oxide
  300.    ref atmos
  200.    lower limit
  600.    upper limit
  160.    current lifetime (yr)
  0.2     dln(lt)/dln(upp-O3)
 -1.0     dln(lt)/dln(circ)

02 B(ppb) CH4              16.05     0.97      2.77 methane
 1600.    ref atmos
  700.    lower limit
 5000.    upper limit
   9.6    current lifetime (yr)    (Prinn et al, 1987)
 -0.95    dln(lt)/dln(trop-OH)      currently strat-loss < 5% of total
  .02     d(loss)/d(trop-T)

03 C(ppm) CO2(C)           12.01     1.00      2.13 carbon dioxide
  345.    ref atmos
  250.    lower limit
  900.    upper limit
  40.     CC carbon constant (yr)
  285.    equ conc/pre-industrial
  -1.     dln(CC)/d(ocean)
   0.     dln(CC)/d(trop-T)                   ?
   0.     dln(CC)/d(CO2)                      ?

04 D(ppt) CFCl3  /11       137.36    0.94      23.0 CFC-11
  220.    ref atmos
    0.    lower limit
 6000.    upper limit
   65.    current lifetime (yr)
  0.2     dln(lt)/dln(upp-O3)
 -1.0     dln(lt)/dln(circ)

05 E(ppt) CF2Cl2 /12       120.91    0.96      20.6 CFC-12
  375.    ref atmos
    0.    lower limit
 6000.    upper limit
  140.    current lifetime (yr)
  0.2     dln(lt)/dln(upp-O3)
 -1.0     dln(lt)/dln(circ)

06 F(ppt) CHF2Cl /22       86.47     0.93      14.3 CFC-22
   80.    ref atmos
    0.    lower limit
 2000.    upper limit
  15.5    current lifetime (yr)    (scale to MC)
 -0.95    dln(lt)/dln(trop-OH)
  .02     d(loss)/d(trop-T)

07 G(ppt) C2F3Cl3          187.37    0.96      32.0 CFC-113
   30.    ref atmos
    0.    lower limit
 2000.    upper limit
   90.    current lifetime (yr)
  0.2     dln(lt)/dln(upp-O3)
 -1.0     dln(lt)/dln(circ)

08 H(ppt) CCl4             153.81    0.93      25.4 carbon tetrachloride
  100.    ref atmos
    0.    lower limit
 1000.    upper limit
   50.    current lifetime (yr)
  0.2     dln(lt)/dln(upp-O3)
```

TABLE 3 (continued)

```
 -1.0     dln(lt)/dln(circ)

09 I(ppt) CH3CCl3       133.40      0.93      22.0 methyl chloroform
  110.    ref atmos
    0.    lower limit
 1000.    upper limit
    6.3   current lifetime (yr)    (Prinn et al, 1987)
 -0.99    dln(lt)/dln(trop-OH)
  .02     d(loss)/d(trop-T)

10 J(ppt) CH3Cl          50.49      0.92      8.25 methyl chloride
  600.    ref atmos
  200.    lower limit
 1200.    upper limit
    1.5   current lifetime (yr)    (scaled to MC)
 -0.99    dln(lt)/dln(trop-OH)
  .015    d(loss)/d(trop-T)

11 K(ppt) CF3Br 1301    148.91      0.96      25.4 halon 1301
    1.7   ref atmos
    0.    lower limit
  100.    upper limit
  110.    current lifetime (yr)
  0.2     dln(lt)/dln(upp-O3)
 -1.0     dln(lt)/dln(circ)

12 L(ppt) CF2ClBr       165.36      0.93      27.3 halon 1211
    1.5   ref atmos
    0.    lower limit
  100.    upper limit
   15.    current lifetime (yr)
  0.5     dln(lt)/dln(col-O3)       (much loss occurs in upper troposphere)
 -1.0     dln(lt)/dln(circ)

13 M(ppt) CH3Br          94.94      0.89      15.0 methyl bromide
   10.    ref atmos
    1.    lower limit
  100.    upper limit
    1.6   current lifetime (yr)    (scaled to MC)
 -0.99    dln(lt)/dln(trop-OH)
  .01     d(loss)/d(trop-T)

14 N(ppt) CF4            88.01      1.00     15.64 carbon tetrafluoride
   60.    ref atmos
    0.    lower limit
 1000.    upper limit
 9999.    current lifetime (yr)

15 T( C ) trop-T             1.         1.        1. tropospheric temperature
  +0.5    1980 pert wrt pre-ind
   -5.    lower limit
   +5.    upper limit
  43.     CT heat constant (W/m2/deg) in yr  CT=tau * 4.3W/m2 / 4 C (2xCO2)
  -1.     dln(CT)/d(ocean)                         (tau = 40 yr)
   0.     dln(CT)/d(trop-T)                  ?

15 steady-state species = X's
 # code   title          at.wt. atm.frac.    Tg/ppb
01 a( % ) trp-OH                                    global avg tropospheric OH
   0.0    ref atmos
  -50.    lower limit
  +99.    upper limit
   0.50   d(trop-OH)/d(NH-OH)
   0.50   d(trop-OH)/d(SH-OH)

02 b( % ) NH-O3                                     N.Hem. tropospheric ozone
   0.0    ref atmos
  -50.    lower limit
 +100.    upper limit
+0.80     dln(NH-O3)/d(col-O3)     <****** possibly = -0.10, because less prod O3 with less UV
```

TABLE 3 (continued)

```
 0.20     dln(NH-O3)/dln(CH4)              (Decreased UV lessen both prod & loss)
 0.10     dln(NH-O3)/dln(NH-CO)
 0.10     dln(NH-O3)/dln(NH/NOx)
 0.10     dln(NH-O3)/dln(NH/NMHC)

03 c( % ) NH-OH                                   N.Hem. hydroxyl radicals
  +0.7    ref atmos offset
  -50.    lower limit
  +99.    upper limit
  +.50    dln(prod)/dln(trop-H2O)  P = J(O'D)*O3*H2O + NOx*HOx
  +.50    dln(prod)/dln(NH-O3)
 -1.00    dln(prod)/dln(col-O3)
  +.10    dln(prod)/dln(NH/NOx)
 0.35     dln(loss)/dln(CH4)       L/OH = k(T)*CH4 + k*CO + k*NMHC + k*OH
 0.40     dln(loss)/dln(CO)         (NH)     35%      40%     15%     10%
 0.15     dln(loss)/dln(NH/NMHC)        sum .LE. 1
 0.10     dln(loss)/dln(OH)2       non-lin.loss (OH+HO2)

04 d(ppb) NH-CO          28.01      0.40     1.99 N.Hem. carbon monoxide
  100.    ref atmos (ppb)
   50.    lower limit
  300.    upper limit
  0.26    nominal lifetime (yr)
 165.     source:  CH4xOH --> CO   (ppb/yr) based on 1600 ppb CH4 & std OH
  40.     source:  NMHCs --> CO    (ppb/yr) scaled to std flux
 200.     source:  emissions       est. 400 Tg in NH (uses flux array instead)
    1.0   dln(loss)/d(NH-OH)       % / %
    1.0   interhem.exch-time(yr)   flux N->S

05 e( % ) SH-O3                                   S.Hem. tropospheric ozone
   0.0    ref atmos
  -50.    lower limit
 +100.    upper limit
+0.80     dln(SH-O3)/d(col-O3)     (%/%) <====== possibly = +0.10, balance between
 0.20     dln(SH-O3)/dln(CH4)                    increased prod & loss, prod less
 0.10     dln(SH-O3)/dln(SH-CO)                  important than in NH.
 0.10     dln(SH-O3)/dln(SH/NOx)
 0.10     dln(SH-O3)/dln(SH/NMHC)

06 f( % ) SH-OH                                   S.Hem. hydroxyl
  -2.2    ref atmos offset
  -50.    lower limit
  +99.    upper limit
  +.50    dln(prod)/dln(trop-H2O)  P = J(O'D)*O3*H2O + NOx*HOx
  +.50    dln(prod)/dln(SH-O3)
 -1.00    dln(prod)/dln(col-O3)
  +.10    dln(prod)/dln(SH/NOx)
 0.50     dln(loss)/dln(CH4)       L/OH = k(T)*CH4 + k*CO + k*NMHC + k*OH
 0.25     dln(loss)/dln(CO)         (SH)     50%      25%     10%     15%
 0.10     dln(loss)/dln(SH/NMHC)        sum .LE. 1
 0.15     dln(loss)/dln(OH)2       non-lin.loss (OH+HO2)

07 g(ppb) SH-CO          28.01      0.40     1.99 S.Hem. carbon monoxide
   60.    ref atmos (ppb)
   30.    lower limit
  200.    upper limit
  0.26    nominal lifetime (yr)
 165.     source:  CH4xOH --> CO   (ppb/yr) based on 1600 ppb CH4 & std OH
  20.     source:  NMHCs --> CO    (ppb/yr) scaled to std flux
  55.     source:  emissions       est. 110 Tg in SH (uses flux array instead)
    1.0   dln(loss)/d(SH-OH)       % / %
    1.0   interhem.exch-time(yr)   flux N->S

08 h( % ) trp-H2O                                 tropospheric water (0-5 km)
   0.0    init pert                 (rel.to trop-To)
  -50.    lower limit
  +50.    upper limit
 0.062    dln(trop-H2O)/d(trop-T)   (vapor pressure at 298K)

09 i( % ) col-O3                                  total ozone column
```

TABLE 3 (continued)

```
   0.0     ref atmos (unused)        std: NOx=18, Clx=0, Brx=0, H2O=5.5, CH4=1600
  -20.     lower limit
   20.     upper limit
   0.0     d(col-O3)/d(upp-O3)       %/%  (already included with the trace gas pert's)
   -1.0    d(col-O3)/d(str-NOy)      %/ppb wrt 18   <====== possibly -0.8, since response to 20%
   -1.0    d(col-O3)/d(str-Clx)      %/ppb                  increase in N2O is too large.
   -0.05   d(col-O3)/d(str-Brx)      %/ppt
   +1.5E-3d(col-O3)/d(CH4)           %/ppb wrt 1600
     .     d(col-O3)/d(str-H2O)      %/ppm wrt 3.0
   +0.     d(col-O3)/d(circ)         %/%
   +0.05   d2(O3)/d(s-NOy)d(Clx)     %/(ppb*ppb)
   -.01    d2(O3)/d(Clx)d(Brx)       %/(ppb*ppt)
   +.20E-3d2(O3)/d(Clx)d(CH4)        %/(ppb*ppb)
   -.03    d2(O3)/d(Clx)2            %/(ppb^2)              /may need to couple CO2 (strat T) wit
   +.01    d(col-O3)/d(CO2)          %/(ppm)        <======  other perturbations (Clx, NOy, CH4)

10 j( % ) upp-O3                                    ozone column above 30 km
   0.0     ref atmos (unused)        NOx=18, Clx=2.78, Brx=12.9, H2O=3, CH4=1600
  -50.     lower limit
   50.     upper limit
   -1.0    d(upp-O3)/d(str-NOx)      %/ppb wrt 18
   -2.5    d(upp-O3)/d(str-Clx)      %/ppb          <======= -4.5, LLNL more sensitive to Clx
   +1.0E-3d(upp-O3)/d(CH4)           %/ppb wrt 1600
   +5.0E-4d2(upp-O3)/d(Clx)d(CH4)    %/(ppb*ppb)
     .     d(upp-O3)/d(str-H2O)      %/ppm wrt 3 ppm
   +.03    d(upp-O3)/d(CO2)          %/ppm wrt 345 ppm

11 k(ppb) str-NOy                                   stratospheric NO-y (35 km)
  18.0     ref atmos                 <======= a better absolute ref is 20 ppb
  12.0     lower limit
  30.0     upper limit
  +.06     d(NOy)/d(N2O)             ppb/ppb   wrt 300 ppb N2O
 -.000067  d2(NOy)/d(N2O)2           ppb/ppb2
   0.      d(NOy)/dln(upp-O3)        ppb/%

12 l(ppb) str-Clx                                   stratospheric Cl-x (40 km)
   2.784   ref atmos
   0.5     lower limit
  18.0     upper limit
  2.95 E-3 d(Clx)/d(CFCl3)
  1.80 E-3 d(Clx)/d(CF2Cl2)           <======= 1.91E-3 choice from LLNL (prefer 35 km as ref alt)
  0.60 E-3 d(Clx)/d(CFC-22)                   0.69E-3
  2.85 E-3 d(Clx)/d(CFC-113)                  2.92E-3
  3.90 E-3 d(Clx)/d(CCl4)                     3.93E-3
  2.95 E-3 d(Clx)/d(CH3CCl3)                  2.80E-3
  0.98 E-3 d(Clx)/d(CH3Cl)

13 m(ppt) str-Brx                                   stratospheric Br-x (25 km)
   12.88   ref atmos
    1.     lower limit
  100.     upper limit
  0.95     d(Brx)/d(CF3Br)
  0.98     d(Brx)/d(CF2ClBr)
  0.98     d(Brx)/d(CH3Br)

14 n(ppm) str-H2O                                   tropopause water
  3.00     ref atmos
   2.0     lower limit
  12.0     upper limit
   0.      d(str-H2O)/d(trop-T)      ? control of tropopause temperature

15 o(W/m2)net-F                                     radiative forcing tropopause
   0.0     pre-ind forcing (for 285=CO2, 700=CH4, 280=N2O, 0=CFCs, 0%=trop-O3)
  -10.     lower limit               ref net-F = 1.55 W/m? (approx)
  +10.     upper limit
 1.075     d(net-F)/d(trop-T)        2xCO2 = 4.3 W/m2 = 1.26 C *f= 4 C --> B=1.075
 3.40      d(net-F)/d(albedo)        W/m2 / % change in albedo
  17.00E-3d(net-F)/d(CO2) ==>        Ts = 4.98E-3(CO2-300) - 2.6E-6(CO2-300)**2
  -8.87E-6d2(net-F)/d(CO2)2             ***exact at 800 ppm, err(1000ppm) <.1
   0.34E-3d(net-F)/d(CH4)            deg ==> W/m2 by   3.4*   = 4.3W/m2 / 1.26C
```

TABLE 3 (continued)

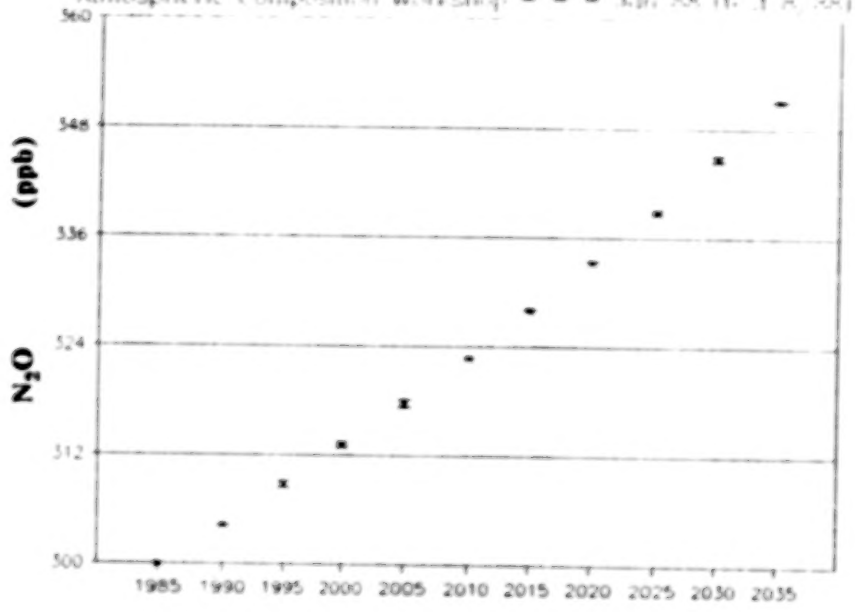
```
 3.4E-3d(net-F)/d(N2O)                    285 --> 300 ppm CO2 = 0.257 W/m2
.224E-3d(net-F)/d(CFC-11)
.286E-3d(net-F)/d(CFC-12)
 .07E-3d(net-F)/d(CFC-22)
 0.     d(net-F)/d(CFC-113)
 .010   d(net-F)/d(trop-O3)       .01 C/D.U., trop-O3=30 DU, 1%=0.3 DU
```

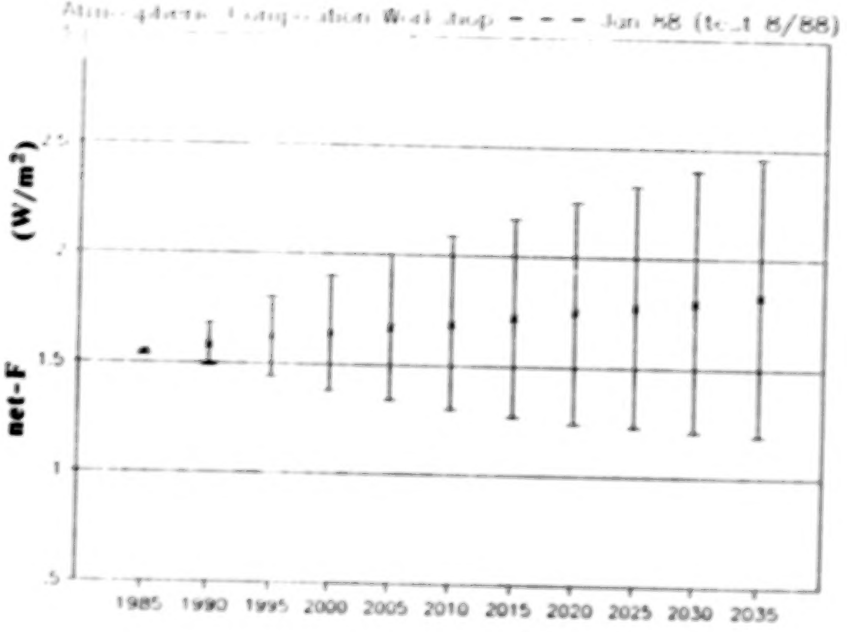
TABLE 4. Input Scenario for Emissions and Coefficient Uncertainties

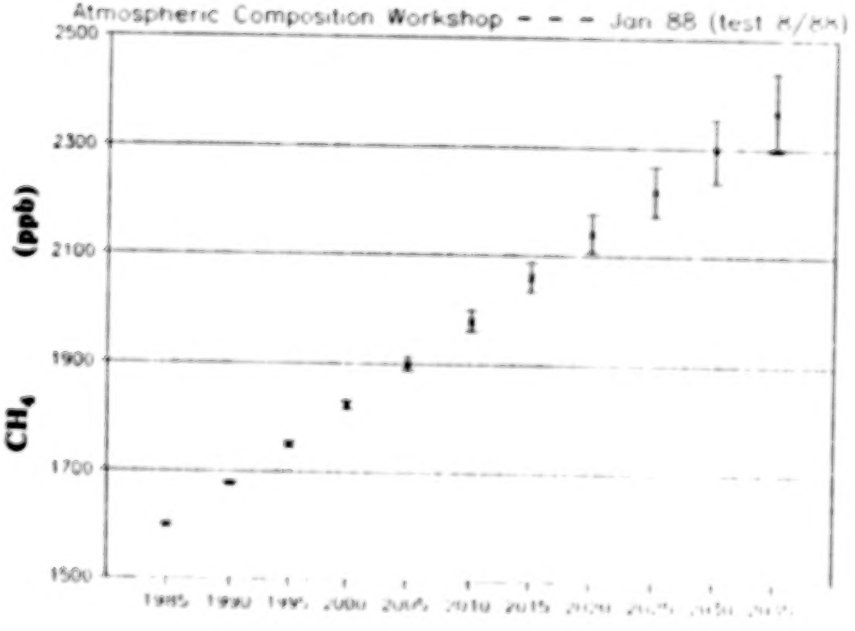
```
 ATCOMP flux 3.0:  based on EPA/LLH combustion scenario & Montreal CFCs UNIT=3
 1985 2060    5 NY0/NY1/NPRTH,        units Xg/yr:  LLH + Montreal
XY title  flux/growth198x 199x  0 (col 11) means Xg/yr, 1 means initial %/yr
01 N2O (N)1     0.25  1.9  -.1  0.9  0.3   .67   .67   .67  0.8  0.8  0.8  0.8  0.8
02 CH4    1      1.0  .76  .79  .78  .75   .60   .60   .60  .27  .27  .27  .27  .27
03 CO2 (C)1      0.4 1.23  .86 1.28 1.46  1.19  1.19  1.19 1.22 1.22 1.22 1.22 1.22
04 CFCl3  0     270.  5.7 -1.5 -0.7 -1.3   0.2   0.5   0.5  0.1  0.1  0.1  0.1  0.1
05 CF2Cl2 0     400.  3.8 -0.5 -0.3 -0.4  -1.9   0.3   0.3  0.1  0.0
06 CHF2Cl 0     225. 10.1  4.9  3.0  2.5   2.5   2.5   2.5  1.2  0.3  0.0
07 C2F3Cl30     140.  0.5 -4.3 -1.1  0.4   0.4   0.4   0.5  0.5  0.5  0.5  0.5  0.5
08 CCl4   1      2.0  3.6 -3.5 -1.0  0.3   0.3   0.4   0.4  0.4  0.4  0.4  0.4  0.4
09 CH3CCl30     520. -3.3  2.7  2.5  2.5   2.5   2.5   2.5  2.5  2.5  2.5  2.5  2.5
10 CH3Cl  1      0.0
11 CF3Br  0       6. 18.0 10.3  6.2  1.8   0.6   0.3   0.3  0.3  0.2  0.0
12 CF2ClBr0       5. 11.2  2.6  2.3  1.0   0.3   0.2   0.5  0.3  0.1  0.1  0.0
13 CH3Br  1      0.0
14 CF4    1      5.0
 0
external scenarios: albedo, ocean, circ, N/S-CO, NOx, NMHCs
01 albedo        0.
02 ocean         0.
03 circ          0.
04 NH-CO        400. -2.4 -2.1 -0.7 -0.9   0.   0.   0.   0.   0.   0.   0.   0.
05 SH-CO         90.
06 NH-NOx        20.   0.   0.   0.  .42  .50  .50  .50  .72  .72  .72  .73  .73
07 SH-NOx        10.
08 NH-NMHC       30.
09 SH-NMHC       10.
 0 ==end==
   3    0    4      Y-species #3  (CO2),   coeff #4
  40.     CC carbon constant (yr)  | time for CO2 uptake
  60.     CC carbon constant (yr)  |
   0   15    4      X-species #15 (net-F), coeff #4
0.750    d(net-F)/d(trop-T)       | climate feedback f = 4.5 = 3.4/B
1.250    d(net-F)/d(trop-T)       |                    f = 2.7
   4    0    4      Y-species #4  (CFCl3), coeff #4
  55.    current lifetime (yr)    | uncertainty in lifetime
  75.    current lifetime (yr)    |
   0    9   14      X-species #9  (col-O3),coeff #14
 -.03   d2(O3)/d(Clx)2            | non-linear response of strat O3
 -.20   d2(O3)/d(Clx)2            |
   0    2    5      X-species #2  (NH-O3), coeff #5
0.20     d(NH-O3)/dln(CH4)        | importance of CH4 in trop
0.40     d(NH-O3)/dln(CH4)        | prod of O3
   0    3    7      X-species #3  (NH-OH), coeff #7
 +.05    dln(prod)/dln(NH/NOx)    | importance of anthropogenic NOx
 +.20    dln(prod)/dln(NH/NOx)    | source for hemispheric OH
   0    0    0 ==end==
```

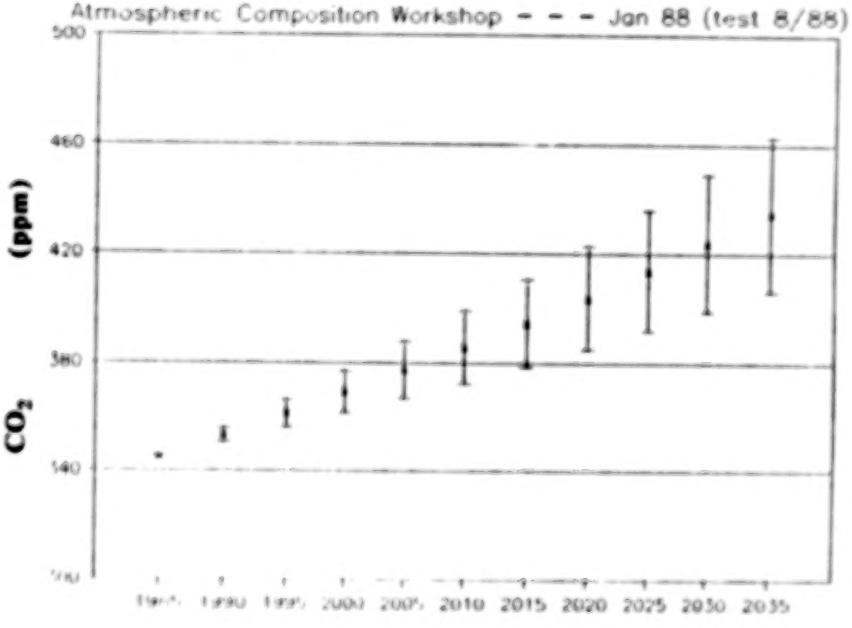
FIGURES (pp. F1-F8)

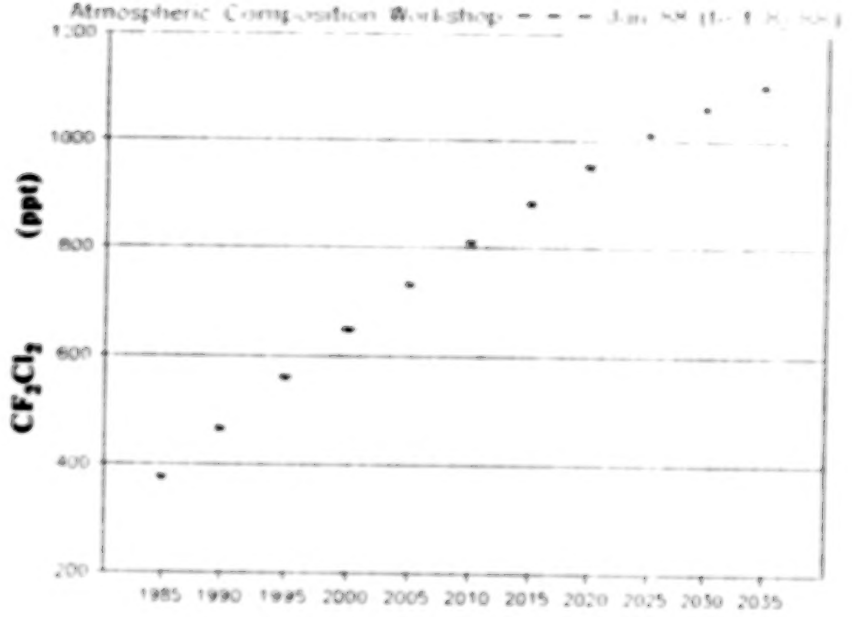
Calculated history of trace gases and climate variables for the assessment model and the emissions scenario described in the text. The center point (X) represents the mean of 2^6 calculations, including all combinations of the ranges specified for 6 model coefficients. The upper and lower limits on each point correspond to ±2 times the r.m.s. variance from the 64 separate predictions.

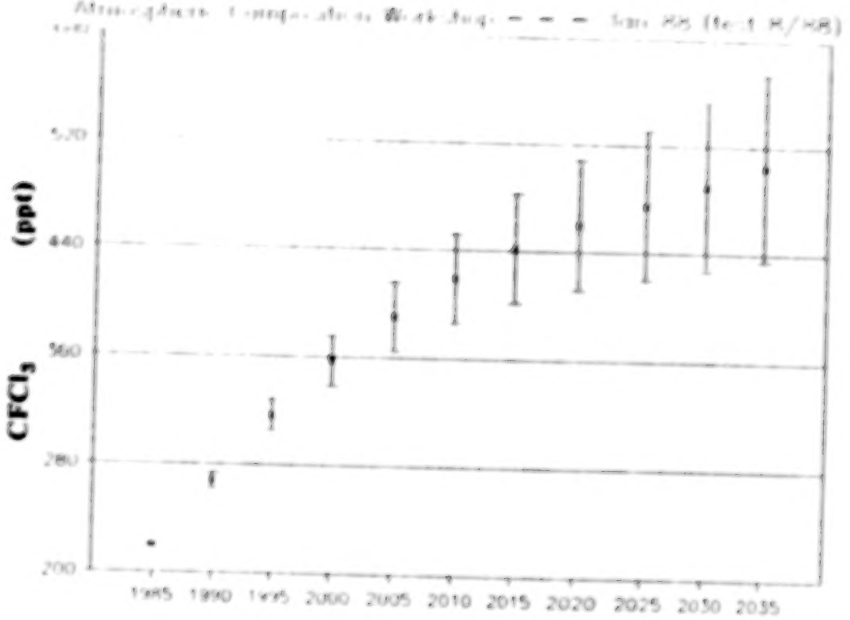

(ppb)
N_2O
560
548
536
524
512
500
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

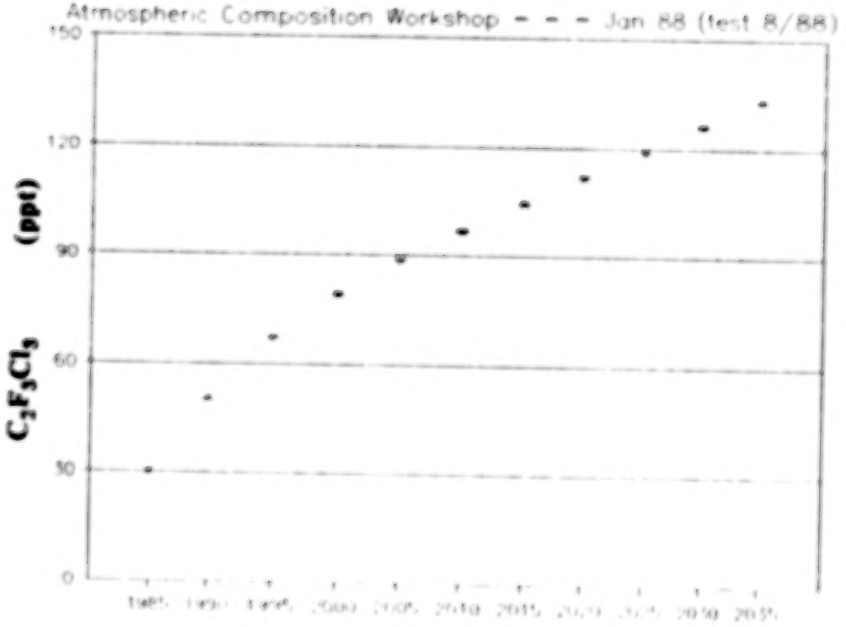

Jan 88 (test 8/88)
(W/m^2)
net-F
2.5
2
1.5
1
.5
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

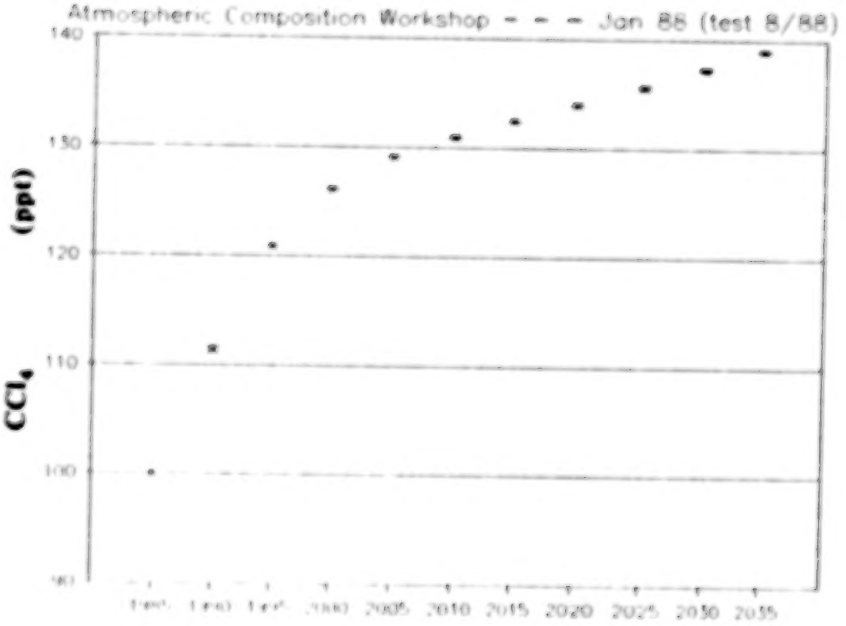

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
(ppb)
CH_4
2500
2300
2100
1900
1700
1500

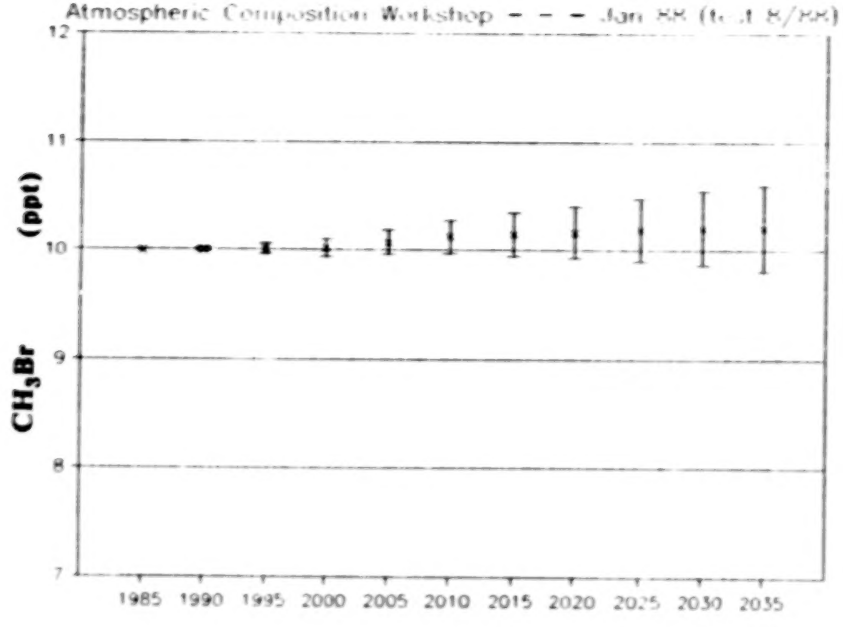

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
(ppm)
CO_2
500
460
420
380
340
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

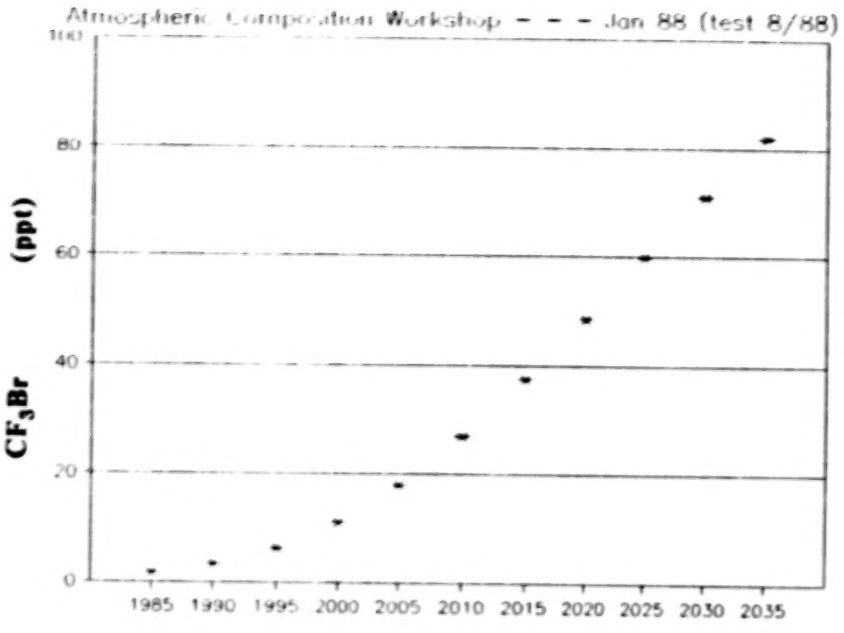

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
CF_2Cl_2 (ppt)
1200
1000
800
600
400
200
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

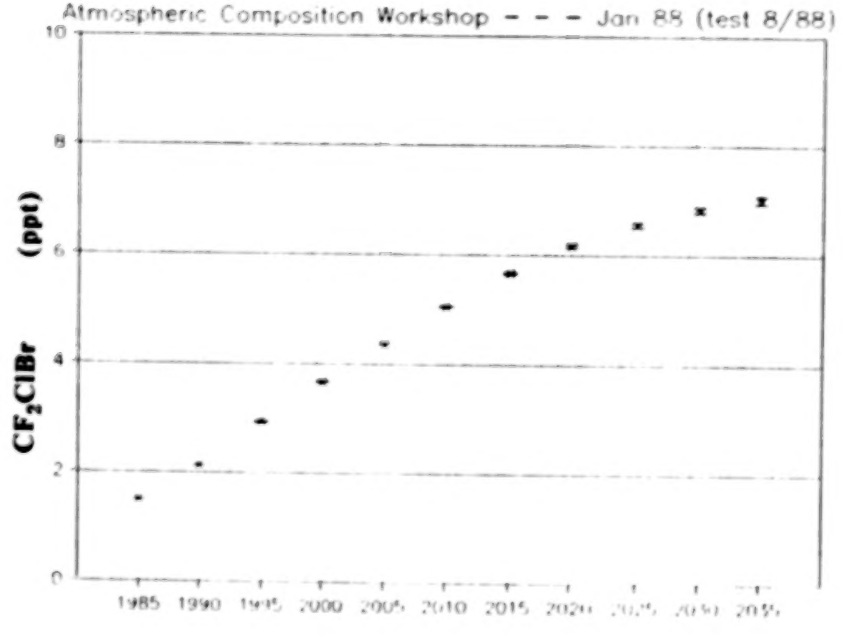

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
$CFCl_3$ (ppt)
520
440
360
280
200
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

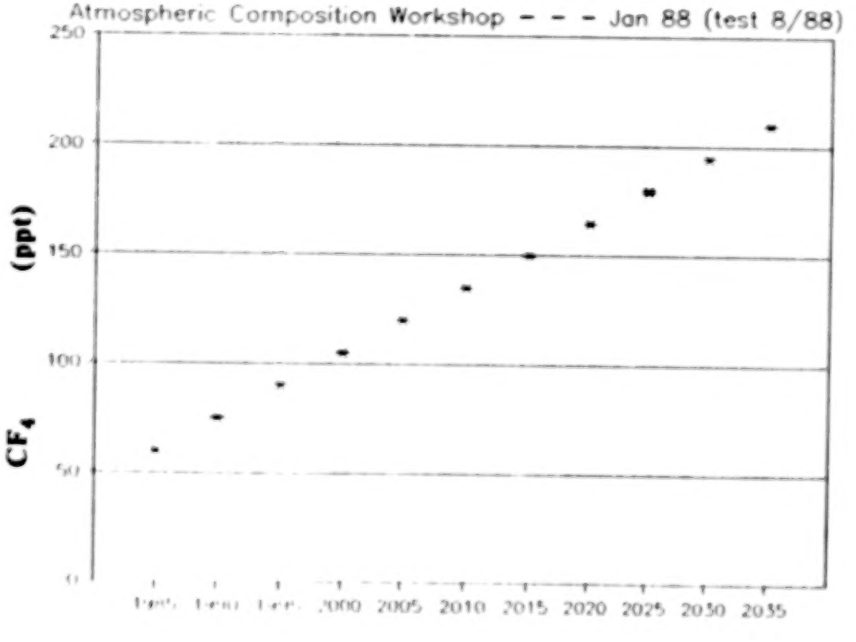

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
$C_2F_3Cl_3$ (ppt)
150
120
90
60
30
0


Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
CCl_4 (ppt)
140
130
120
110
100
90
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

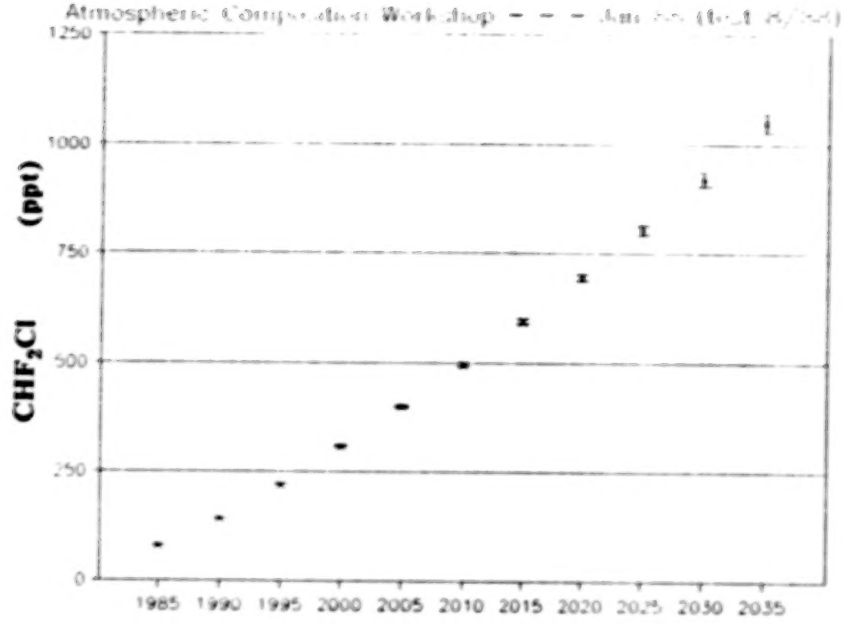

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
CH_3Br
(ppt)
12
11
10
9
8
7
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

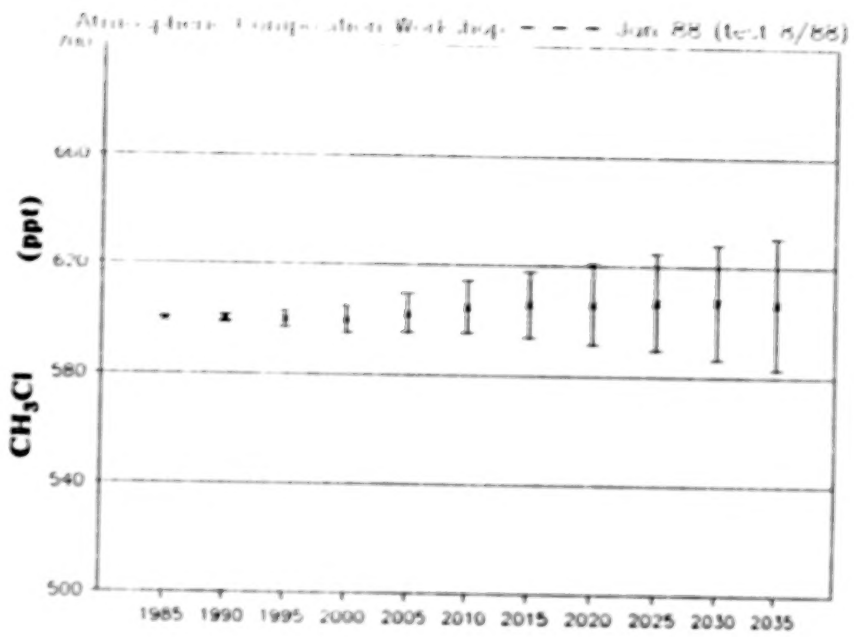

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
CF_3Br
(ppt)
80
60
40
20
0
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

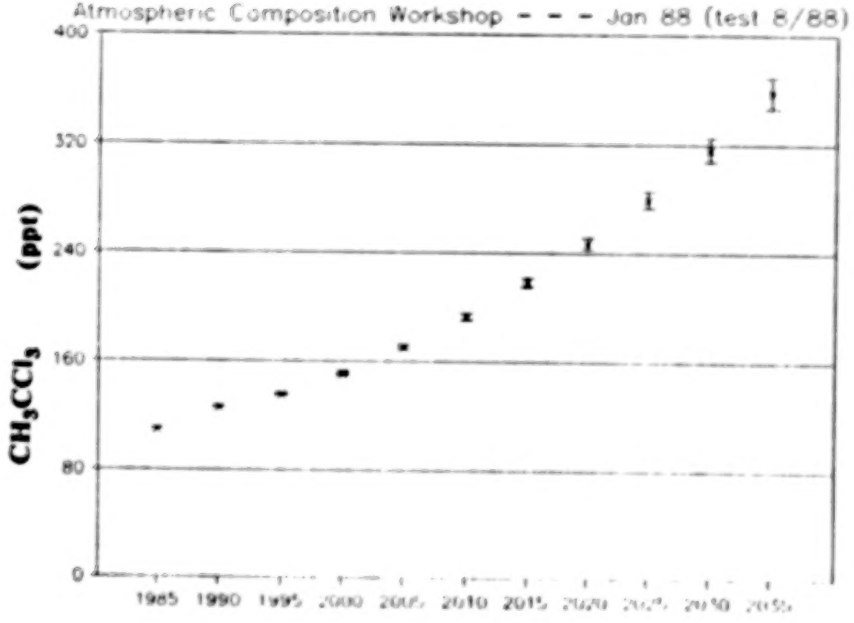

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
CF_2ClBr
(ppt)
10
8
6
4
2
0
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

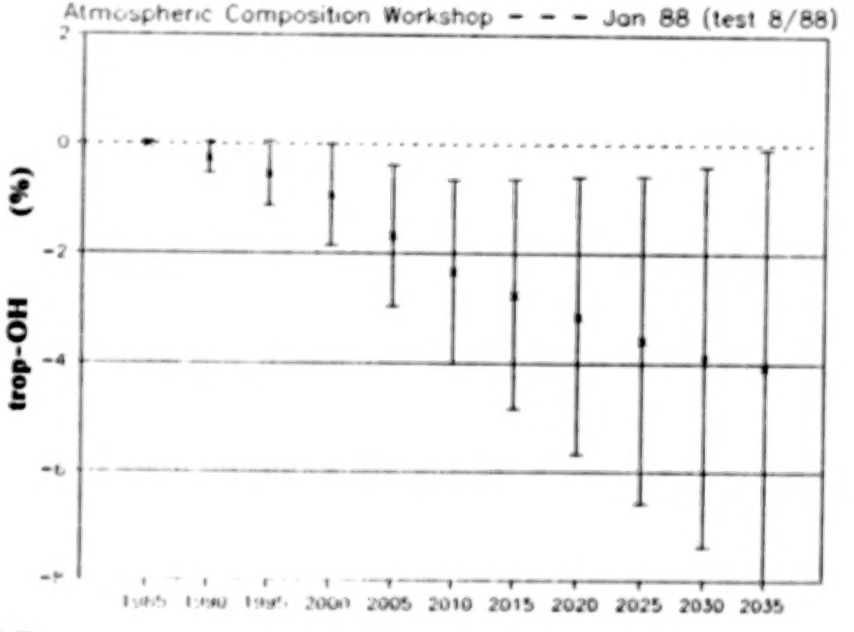

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
CF_4
(ppt)
250
200
150
100
50
0
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

F3

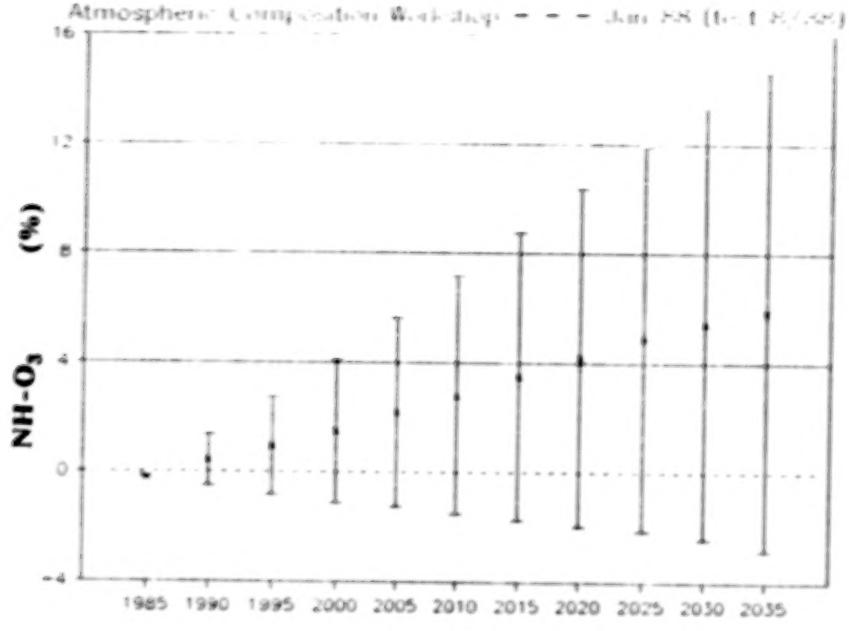

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
CHF_2Cl (ppt)
1250
1000
750
500
250
0
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

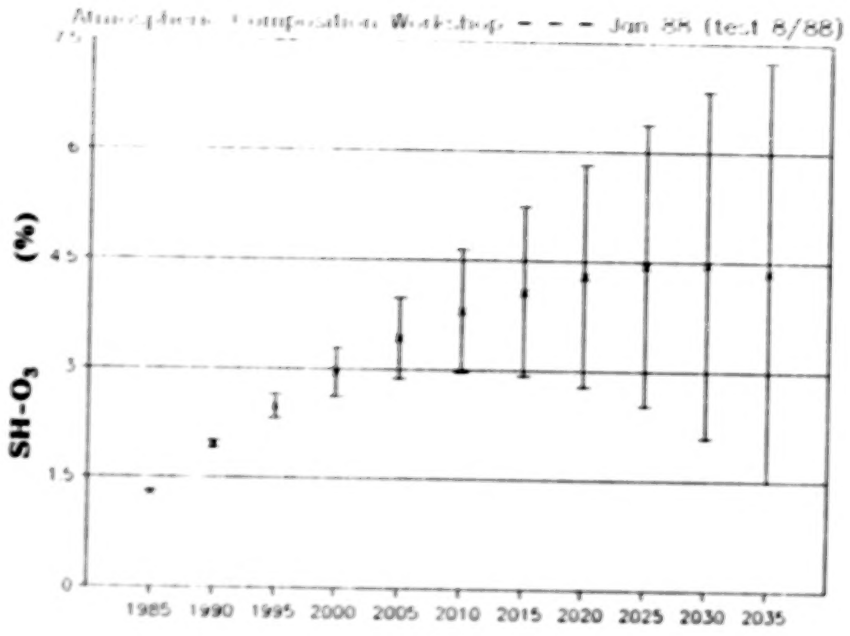

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
CH_3Cl (ppt)
640
620
580
540
500
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

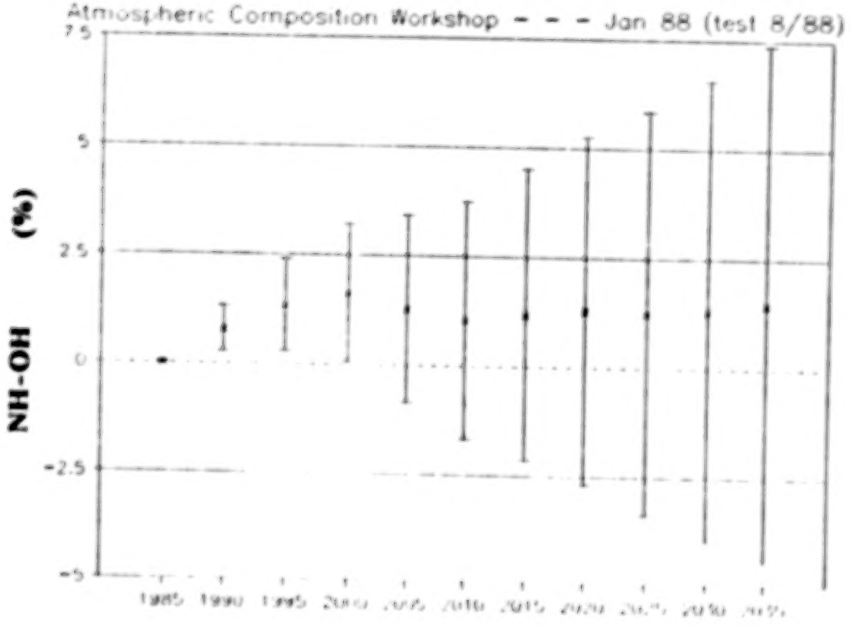

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
CH_3CCl_3 (ppt)
400
320
240
160
80
0
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

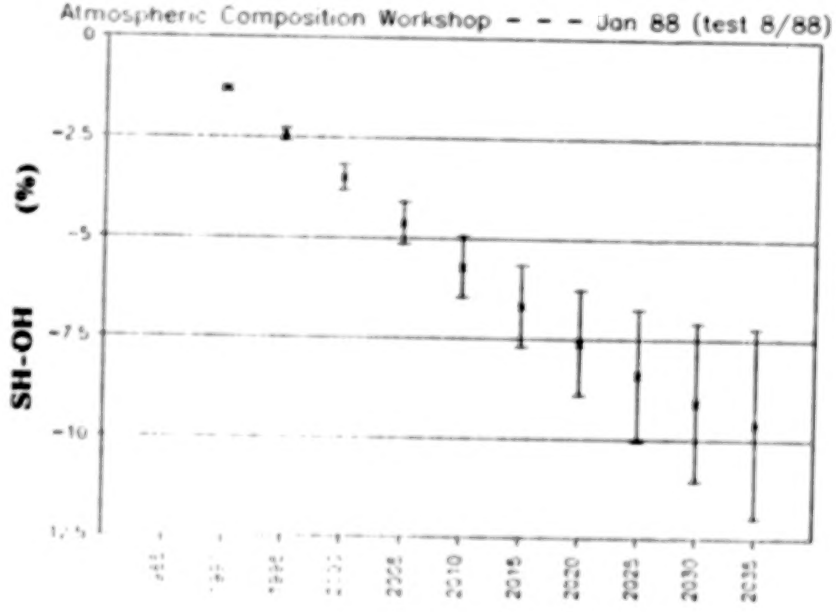

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
trop-OH (%)
2
0
-2
-4
-6
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

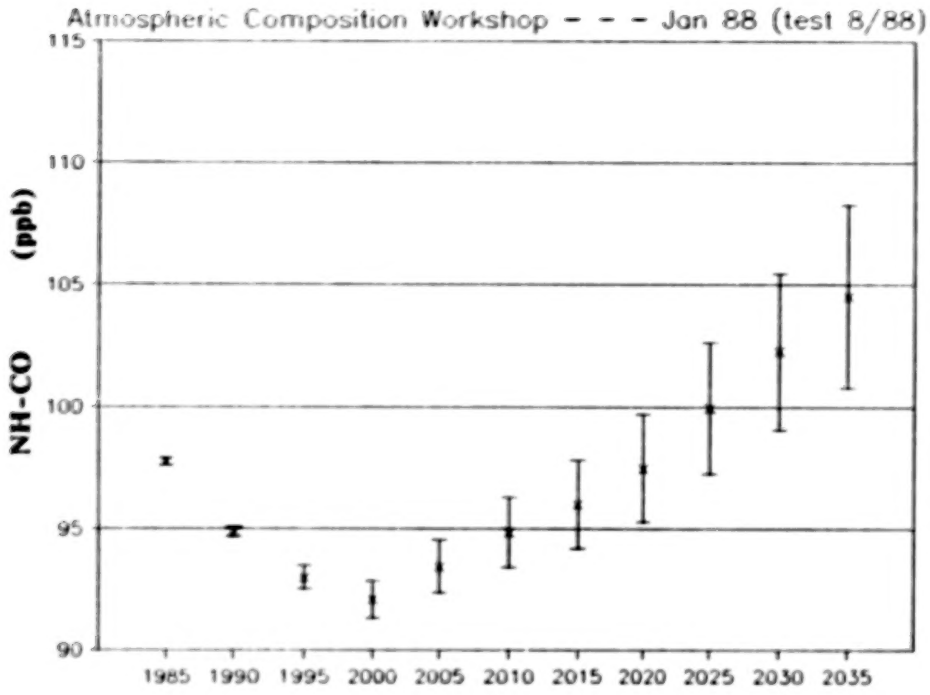
F4

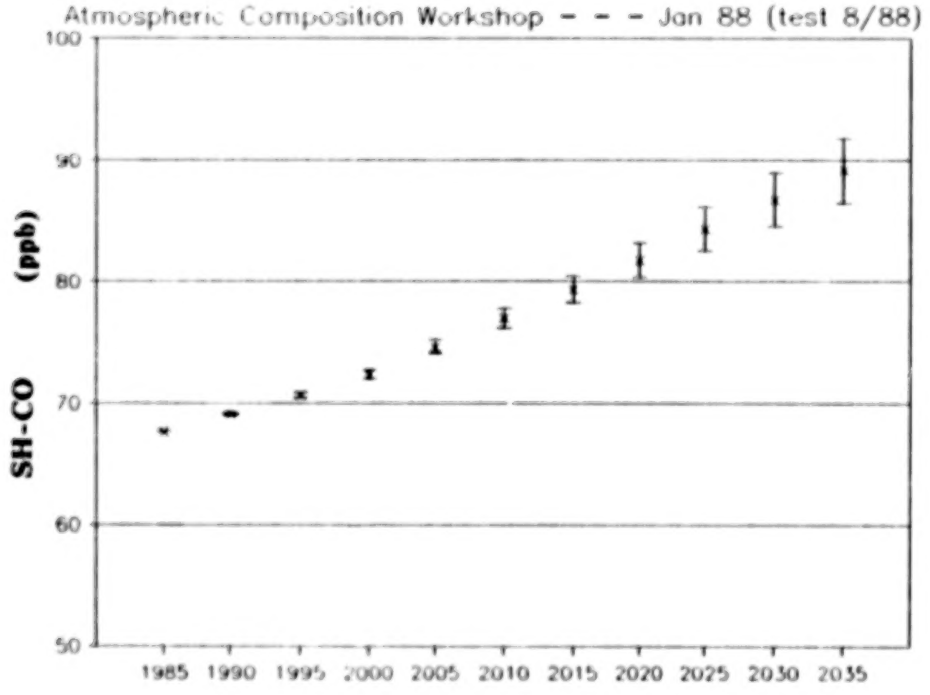

NH-O3 (%)
16
12
8
4
0
-4
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

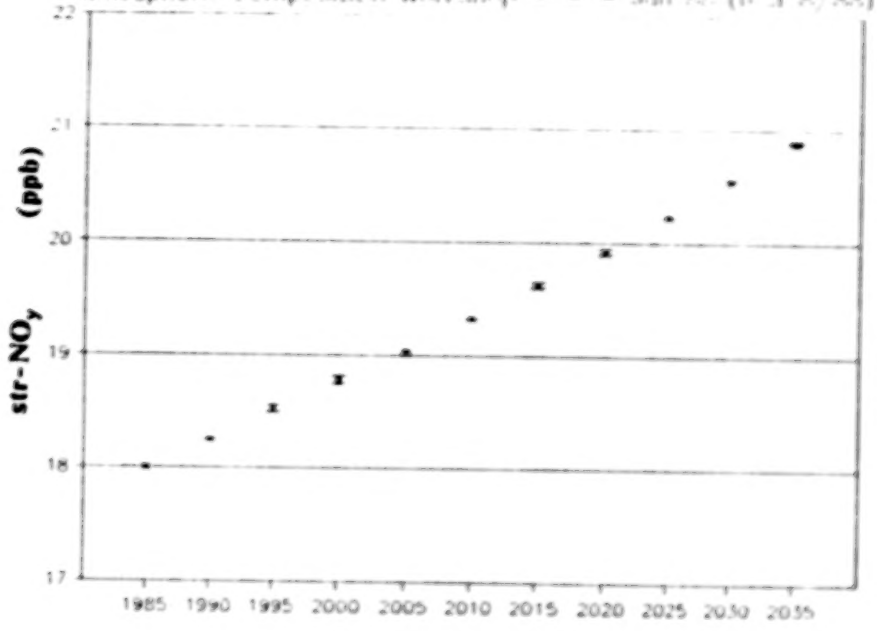

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
SH-O3 (%)
6
4.5
3
1.5
0
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

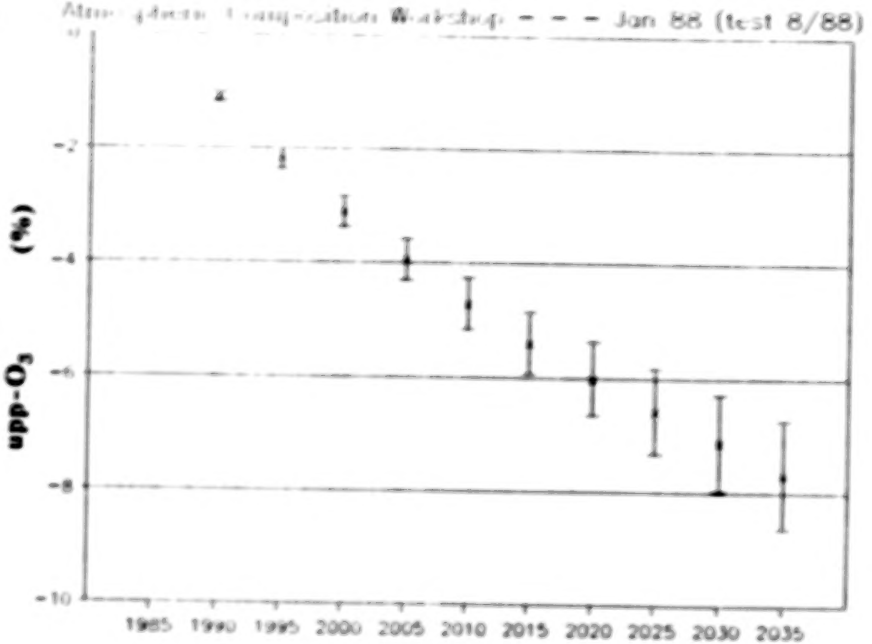

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
NH-OH (%)
7.5
5
2.5
0
-2.5
-5
1985 1990 1995

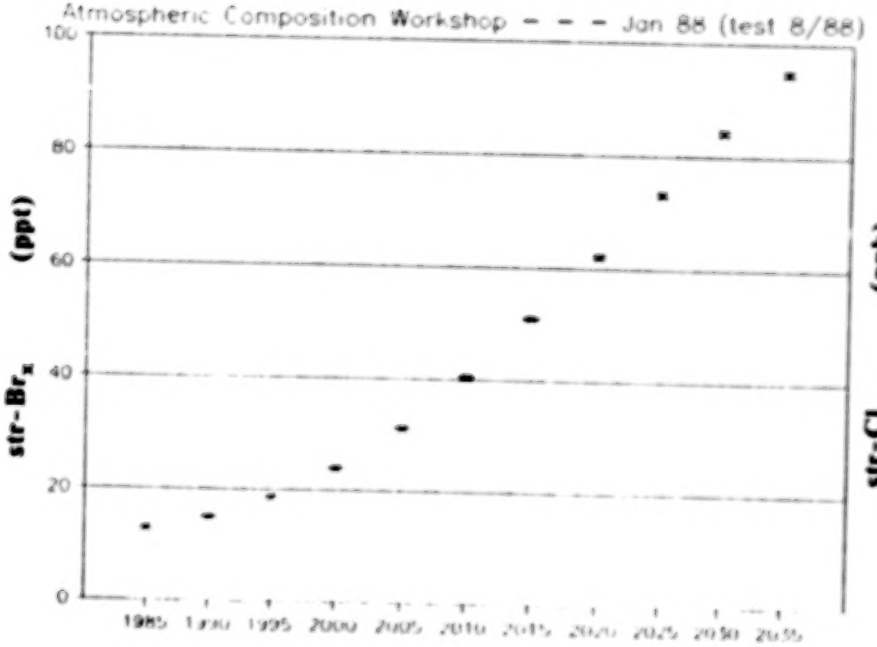

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
SH-OH (%)
0
-2.5
-5
-7.5
-10
2010
2015
2020
2025
2030
2035

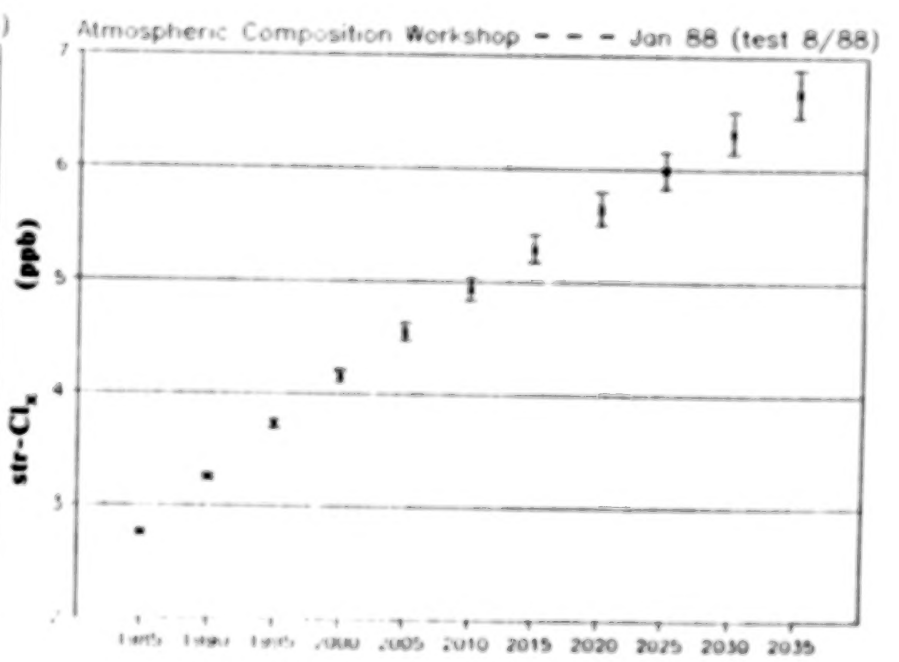

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
NH-CO (ppb)
115
110
105
100
95
90
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035


Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
SH-CO (ppb)
100
90
80
70
60
50
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

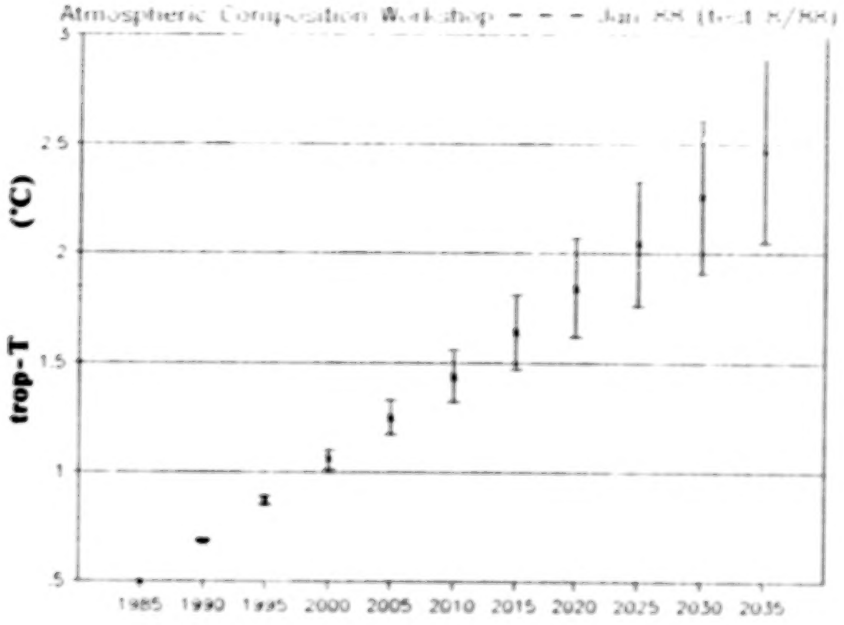

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
str-NO$_y$ (ppb)
22
21
20
19
18
17
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

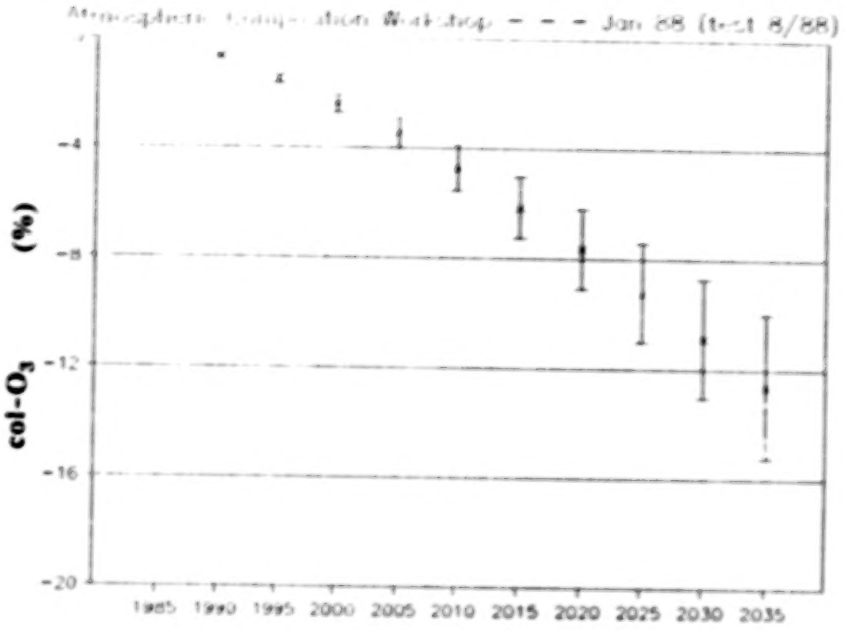

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
upp-O$_3$ (%)
-2
-4
-6
-8
-10
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035


Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
str-Br$_x$ (ppt)
100
80
60
40
20
0
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

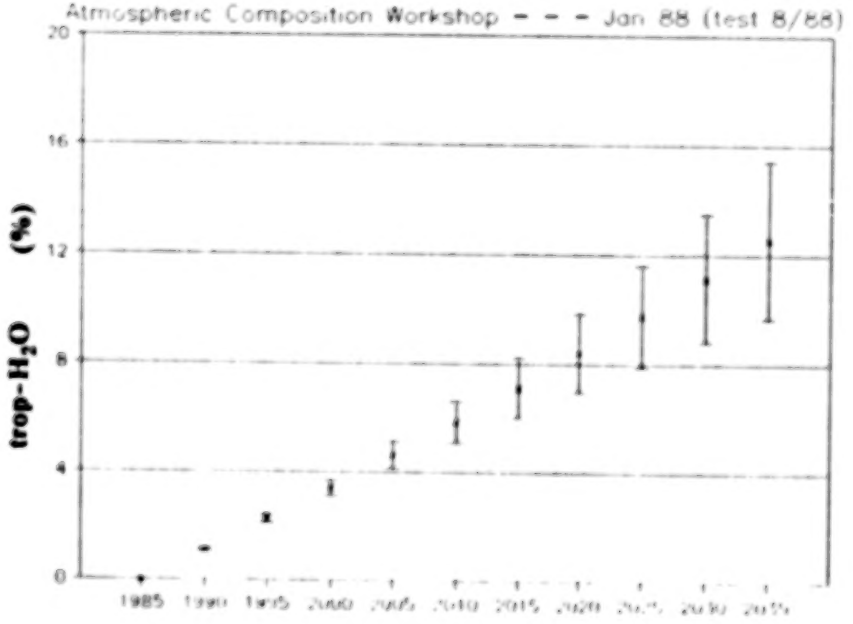

Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
str-Cl$_x$ (ppb)
7
6
5
4
3
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

F7


trop-T (°C)
2.5
2
1.5
1
.5
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035


col-O3 (%)
-4
-8
-12
-16
-20
1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035

8d


Atmospheric Composition Workshop - - - Jan 88 (test 8/88)
trop-H2O (%)
20
16
12
8
4
0
1985

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15. Supplementary Notes

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Michael J. Prather - NASA/GISS, 2880 Broadway, New York, New York.

16. Abstract

Predicting future perturbations to global air quality and climate requires, as a prerequisite, prognostic models for the composition of the Earth's atmosphere. Such assessment models are needed to evaluate the impact on our environment of different social choices that affect emissions of the photochemically and radiatively important trace gases. Our presentation here of a prototype assessment model is intended to encourage public scientific discussions of the necessary components of the model and their interactions, with the recognition that models similar to this will likely be used by the Environmental Protection Agency and other regulatory agencies in order to assess the effect of changes in atmospheric composition on climate over the next century.

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